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Transparency Is a Term Often Used But Hard to Define

I am sure you have been in this situation: You're in someone's office and while you wait for them to finish a telephone conversation, you read a potpourri of sayings, plaques and pictures hanging on the walls and find one that catches your eye and makes you think and sometimes smile. Here is one that I thought summed up various aspects of the materials management profession we represent: *Predicting the future is not really that difficult; the challenge that faces us is determining what's happening today.* This is reflective of the domestic and international challenges that we face today as we attempt to understand the full impact of the end of the Cold War.

The International Atomic Energy Agency (IAEA) held its 1994 Symposium on International Safeguards in Vienna, Austria, March 14–18. The INMM was a cooperative organization, along with the American Nuclear Society, the European Safeguards Research and Development Association, and the Russian Nuclear Society. The symposium featured approximately 200 papers (oral and poster), with an attendance of approximately 400 people.

Its focus was excellent and very timely for discussing the issues facing the IAEA. Although there were discussions about improving the efficiency and effectiveness of classical IAEA safeguards, there were a considerable number of papers on expanding the IAEA role of detecting clandestine or undeclared activities. The situations in Iraq, North Korea and elsewhere prompted these discussions. Terms used freely included *inter alia*, special and unannounced inspections, environmental monitoring, remote monitoring, transparency and openness. Also discussed were new areas that could potentially involve the IAEA, such as the disposition of released plutonium (*released* referring to plutonium from weapons programs), the Comprehensive Test Ban Treaty and the proposed SNM Cut-Off Treaty. There certainly was no unanimous agreement among the participants on these topics, but the symposium offered the opportunity for discussion. The mood of the participants was pragmatic and often nonparochial in addressing approaches to solving the worldwide horizontal and vertical proliferation problems.

James Larrimore, the scientific secretary of the IAEA and INMM Vienna Chapter chair, did an excellent job in arranging the program.

On March 18, immediately following the symposium, the International Safeguards division of the INMM held a timely meeting on transparency. This is a term that is used quite frequently nowadays, yet it has a different meaning for different people and can be an emotional issue. During the symposium, there were presentations that defined transparency as "additional information," which could allow judgment to be passed regarding nondeclared operations. There were some who opined that transparency was additional information plus openness, implying anywhere, anytime inspections. I was left with the impression that when the international community speaks of transparency, it is addressing the problem of undeclared or clandestine activities. This is perhaps different from the use of the word in the United States, where generally it relates to confidence-building and a nonintrusive approach to verification issues.

During the International Safeguards division meeting, attended by 52 experts from the community, we attempted to clarify the meaning of transparency or at least come to a

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B. Pellaud (third from the right), deputy director general for safeguards, acknowledges the cooperation of the supporting professional societies for the 1994 IAEA Symposium on International Safeguards in Vienna, Austria, March 14–18. The organization representatives are (from left): J. Larrimore, IAEA scientific secretary and INMM Vienna Chapter chair; A. Carnino, American Nuclear Society; M. Cuypers, ESARDA; Pellaud; D. Mangan, INMM; and V. Shmelev, Russian Nuclear Society.

Management of Spent Fuel Is a Big Business

In 1982, the U.S. Congress decreed that the Department of Energy (DOE) take possession of and permanently dispose of the spent fuel and highly radioactive wastes generated by civilian nuclear activities. Every January since then, the INMM sponsors a spent fuel management seminar in Washington, D.C. Since the management of spent fuel is a big business, there are other, much larger symposia. Our seminars are so wisely planned by Division Chair E.R. Johnson and his division members that one can learn about everything that is going on in two-and-one-half days of single sessions. Most contributors provided copies of their papers or viewgraphs, which are available from INMM headquarters, 60 Revere Dr., Suite 500, Northbrook, IL 60062, tel. 708/480-9573, fax 708/480-9282.

The major emphasis for the electric utilities and the responsible government agencies is on how to implement the storage and disposal program. Many of the nuclear reactor pools are so full that the utilities must consolidate fuel assemblies or construct dry storage facilities on the site. The DOE Office of Civilian Radioactive Waste Management is investigating a possible repository site and, at the same time, it must reach agreement with the utilities on how the fuel assemblies are to be arranged in storage containers and can be used with the DOE's transportation and disposal overpacks when the spent fuel is picked up. Until this year, there was little discussion regarding the application of International Atomic Energy Agency (IAEA) safeguards. In the final session of the Spent Fuel Management Seminar, William Murphy from the Nuclear Regulatory Commission discussed the need to support IAEA safeguards for spent fuel, as was laid out in the U.S./IAEA voluntary agreement.

We have the pleasure of publishing a short paper by Pierre Saverot regarding the definitions of and regulations for low-level radioactive waste disposal of several major nations. At the January Spent Fuel Seminar, Saverot said France requires all spent fuel assemblies be measured nondestructively at the reactor before being shipped and at a reprocessing plant before being chopped up for dissolution. He also described three of the NDA systems which were designed and tested for this application.

This issue also features three more papers: one from the Spent Fuel Seminar, one for chemists, on whom all our measurements depend, and one for the NDA entrepreneurs.

This issue also has a letter to the editor and a response. I hope that we will have more in the future. Regarding the question of what to do with the plutonium, I observe that nothing is going to be buried in Yucca Mountain until at least 2010, and that any such items are to be retrievable for another 50 years. By 2060, it is likely that fast breeders will be very attractive economically and environmentally.

Recently, the INMM headquarters received a copy of a report that may be of interest: "Security Issues in the Handling and Disposition of Fissionable Materials," by Herbert L. Abrams and Dan Pollak. First, they present a reasonable description and critique of the DOE's physical security and material accounting and control program for fissionable materials. Second, they observe that the former Soviet Union will be dismantling nuclear warheads and storing or processing the fissionable materials but lack a comparable material control and accounting and physical security system. Finally, they appeal to the United States and other countries to assist the Russians in the urgent and difficult process of designing and implementing a credible protection system for the materials. Some of you may be interested in the authors' description of the DOE system. Others probably are involved in attempting to assist Russia in this area. Copies may be obtained from the authors at the Center for International Security and Arms Control, 320 Galvez St., Stanford, CA 94305-6165.

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Additional Thoughts On What We Should Do With the Plutonium

I would like to add some thoughts on "What Should We Do With the Plutonium," John Bartlett's comment in the January 1994 *JNMM*. I hope we hear more about this very important topic from other readers.

For more than a decade, numerous individuals and institutions studied the subject of surplus plutonium disposal, and there is a plethora of opinions on the subject. The most recent studies were by the General Accounting Office on transmutation, the Office of Science and Technology Policy and the National Academy of Sciences (NAS). In addition, there have been ongoing discussions at numerous professional society forums.

In spite of all these studies, Bartlett's comments lack a fundamental recognition of some realities of our time. At a minimum, we have to recognize the following before we start pontificating on the destruction of plutonium:

- Plutonium was discovered in 1940 by Arthur Wahl, John Kennedy and Glen Seaborg. If they had not done so, someone else would have. Plutonium cannot be disinvented. Plutonium exists, and we must learn to live with it.
- For the past 50 years, the science and technology for producing, fabricating, storing, safeguarding and using plutonium was well-developed, and that knowledge cannot be erased.
- There is approximately 1,500 Mt of plutonium now in the world, including plutonium contained in spent fuels, and that inventory is increasing at the rate of about 60 Mt per year.
- The amount of plutonium that will likely emerge from weapons dismantlement (if the unilateral declarations by the United States and Russia and bilateral treaties are ratified and executed) is less than 100 Mt. The titles to this excess plutonium will remain with the United States and Russia, the original owners.
- The United States and Russia are not the proliferators of this world. The excess plutonium removed under treaty agreements in their custody will require monitoring to prevent surprises. The present unsettled political situation in Russia is of greater concern than the potential of a surplus plutonium inventory in their possession. There is no chance that Russians will hurry up, dismantle their weapons and destroy the plutonium to satisfy the fantasies of people.
- Plutonium still has a role to play in the national security strategies of several weapons states. That is not likely to change in the near future.

Over the years, people recommended sending excess plutonium to outer space, using it in a big bang, and burning it in all kinds of contrived new devices that will create economic activity. Most recently, the NAS felt that it, too, must add to the ridiculous by proposing to bury excess plutonium in a 2.5-mile-deep hole. Although this scenario was highly publicized in the press, the Academy's first recommendation — burning the surplus plutonium in existing reactor designs — is the most sensible proposed so far.

Burning plutonium in existing reactor designs was proposed by many people in the past, and it is good to know that the NAS endorses that scenario. Also, there were other sensible suggestions ranging from long-term storage to burning plutonium in several new reactor designs. I am sure INMM members will recognize that the NAS's proposal to mix excess plutonium in vitrified, high-level wastes would be a safeguards nightmare.

There is an almost completed MOX fuel fabrication facility at Hanford, paid for by U.S. taxpayers, that can be readily modified to fabricate the excess weapons plutonium into usable fuel. Adequate MOX fuel fabrication facilities and reactor types exist in the former Soviet Union to burn not only their plutonium surplus but probably that of the United States, too. Such fabricated fuel can be burned in almost all existing U.S. reactors and several reactors in Japan and Europe under IAEA safeguards. There is no need for any new inventions to carry out this scenario.

Adding spent fuels from this scenario to the existing inventories of spent fuels in the world would have only a minuscule impact. Such consumption of excess plutonium for energy production will remove the excess plutonium inventory in a short time and will generate useful energy, which the world badly needs.

There are approximately 30 nations with nuclear power programs and 14 with various levels of reprocessing capabilities. President Clinton's Sept. 20, 1993, nonproliferation initiative is a highly desirable goal. However, the realists of this world will recognize that it will be a long while before such a consensus can be developed worldwide. The nuclear weapons states, other than the United States and the former Soviet Union, have not yet spoken about reductions in their arsenals. In fact, some were testing and improving their weapons while the United States and the former Soviet Union were competing with each other in announcing voluntary reductions and negotiating bilateral and multilateral (with the new states of the former of Soviet Union) agreements to reduce their nuclear arsenals.

I hope *JNMM* readers will not contribute to creating a feeling of impending doom because a few tons of plutonium are likely to be removed from weapons inventory.

K.K.S. Pillay
Los Alamos, New Mexico U.S.A.

John Bartlett Responds

I agree with K.K.S. Pillay that burning surplus weapons-grade plutonium in existing reactor designs would be a sensible method for disposition of this material, and I suspect that many in the relevant technical communities share this view.

However, many others with an interest or role in this issue do not share this view. They are predominantly concerned with the potential threat of the plutonium and the threat of re-institutions of nuclear weapons, terrorist theft and toxic effects on human health. These threats will exist as long as the plutonium exists, and elimination of the plutonium from existence in the human environment is, therefore, a priority goal for those people. To accomplish this goal, whatever the means, will take some time, so there is near-term emphasis on physical security and internation equity in management and disposition of inventories. A companion goal is to cease the expansion of plutonium inventories that results from nuclear power operations and spent fuel recycling.

Potential means of plutonium disposition were identified but not well characterized. The basic options are the same as those for disposal of fission reactor wastes: isolation from the human environment in geologic formations, destruction by nuclear reaction or elimination from earth by ejection to space. All of the options are technically feasible, but each has associated environmental, economic, security and social characteristics, and radiation exposure potential that were not evaluated.

My article was a plea for total-system characterization of the disposition options as a basis for choice of policy and action. I also tried to point out that direct disposal of weapons-grade plutonium would have numerous, diverse, significant and adverse impacts on the present U.S. program for disposal of commercial spent fuel. I elaborated these impacts in a paper titled "The Impacts of Weapons-Grade Plutonium Disposal on the Spent Fuel Management System," presented at the INMM Spent Fuel Seminar XI, held in Washington, D.C., Jan. 26-28, 1994.

A balanced, comprehensive assessment of the options for disposition of weapons-grade plutonium may well not be the basis for policy and action. The sensible thing to do depends on the sensibilities of those in charge. At present, as stated by Wolfgang Panofsky, chair of the National Academy of Sciences plutonium study group (quoted in *Science*, volume 263, Feb. 4, 1994, page 631), "Maintaining the plutonium under full national and international control and preventing its distribution and theft are the main priorities. The name of the game is security, not economics."

Seminar Shows Progress in Spent Fuel Storage and Disposal

The 11th INMM Spent Fuel Management Seminar was held Jan. 26-28 in Washington, D.C. The backdrop to this seminar included the recent appointments and confirmations of Daniel Dreyfus as the new director of the Department of Energy's (DOE) Office of Civilian Radioactive Waste Management (OCRWM), and Richard Stallings as the new U.S. nuclear waste negotiator.

Dreyfus was the opening speaker of the seminar. After describing the technical and management progress that were achieved, he reiterated the DOE's commitment to pursue the completion of the characterization of Yucca Mountain in a timely and aggressive manner. Dreyfus stated that it is important to implement policies which eliminate from future generations the burden of disposing of spent nuclear fuel.

Stallings was the luncheon speaker, and he used the recent winter storm and the associated power shortages as an example of the continued need for nuclear power in the United States. It is his position that a monitored retrievable storage facility (MRS) is indeed needed. Stallings added Robert Liimatainen to his staff to provide expertise in science and technology. Because of the national focus of the negotiator's office, Stallings moved the media relations activities to the Washington, D.C., office and hired Maureen Conley, former editor of *Radioactive Exchange,* to run the office. Stallings believes that a possible way to get the process moving is to make the MRS part of a high-tech science park so that communities will host not just a nuclear waste site but an economically valuable research project.

Stallings also said that the negotiator's office closes in January 1995. Several localities already contacted his office to express their interest, including four Native American tribes. Stallings said that if the MRS is not sited voluntarily, Congress will reach a crisis point and force a solution by imposing the MRS somewhere on a federal installation. Stallings encouraged the Mescalero Apache tribe to continue pursuing the private MRS route, thus breaking new ground that might be the solution to the problem.

Fred Peso, vice president of the tribe, also participated in the seminar. Peso stated that although the tribe remains an applicant in the voluntary MRS siting process, it is frustrated by the delays and lack of progress toward a resolution of the siting issue. The council is still open to negotiations with the federal government on a MRS siting agreement. The Mescalero Apache tribe views the operation of an MRS as a tremendous business opportunity, with or without federal involvement. Although the tribe remains an active participant in the voluntary siting process, until a new process is established and funding becomes available, it will also explore the possibility of siting a private, commercial sector MRS.

Steven Kraft of the Edison Electric Institute stated in his presentation that the new administration's appointment of "an experienced and capable team of leaders" indicates a "strong commitment to achieving a resolution to the spent fuel management issue." Kraft reported that utilities remain concerned about sustaining or even accelerating progress in the site characterization project, and that the key element for achieving this is adequate and predictable funding for the program.

Judith Pensabene of the Senate Energy and Natural Resources Committee suggested there should be a separation of the issues of future nuclear power generation and nuclear waste disposal. She said that nuclear waste disposal should be approached as an environmental issue, addressing both interim storage and ultimate disposal.

Dennis Price of the U.S. Nuclear Waste Technical Review Board (NWTRB) presented an entirely different perspective. He said that the OCRWM program is driven by "unrealistic scheduling" and additional study is necessary. He stated that the DOE's approach to the development of a multipurpose canister (MPC) is an example of hasty decision making, and he believes that the impetus behind this is the pressure on the DOE to comply with the 1998 date set forth in the standard contract for spent fuel acceptance. Price concluded by saying that the NWTRB believes that "making decisions based principally on a desire to meet arbitrary schedule deadlines may undermine the technical and scientific underpinnings of those decisions and could actually delay the program or make it more expensive."

John Jicha of the DOE delivered a presentation on the DOE Spent Nuclear Fuel Program. He reported that the DOE decided it will not reprocess its spent fuel in the future and that this fuel should be included for disposal in the first repository. Included in the scope of the program are the tasks of identifying, inventorying, characterizing, stabilizing, and treating (as necessary) the more than 90 different types of DOE spent fuel for disposal in the national geologic repository.

Jean-Claude Guais of NUSYS presented the French view of the benefits of reprocessing and the use of plutonium fuels, and provided an analysis of the economic justification for their use. He reported that the recycling of spent uranium fuel, with its subsequent remanufacture into MOX fuel, could, under favorable conditions, produce a savings in fuel procurement expenses of approximately $14 million

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per annual reload for a 900 MWe PWR.

Ferenc Takáts of the International Atomic Energy Agency (IAEA) provided an overview of the international status and trends of spent fuel management. He also highlighted future IAEA activities, including the preparation of international guidelines on the safety of spent fuel storage and advisory programs on spent fuel management safety. [Takáts' paper on this subject is on page 40.]

Aurelio Ulibarri of ENRESA presented a paper on the Spanish high-level radioactive waste management system, its mission, development, structure, funding, strategies and systems design objectives. This system will include reprocessing and interim storage until a deep geologic repository is available for final disposal of the high-level waste. The increase in necessary interim storage capacity will be provided by three methods: pool reracking, metal storage casks including dual-purpose casks, and the construction of a centralized interim storage facility.

Additional sessions included papers on burnup credit; its verification using the FORK measurement system; its use in storage, transportation, and repository disposal; impacts on utility operations; and burnup measurement systems used in France. Presentations were also made on the current projections on U.S. utility out-of-pool storage requirements, the current status of dry spent fuel storage licensing activities by the Nuclear Regulatory Commission, the impacts of weapons-grade plutonium disposal on the spent fuel management system, and IAEA safeguards for spent fuel in storage and repository disposal.

The status of current storage technologies was well-described in presentations from both the vendor and utility-user perspectives. There were papers on vertical concrete cask storage, concrete vaults, horizontal concrete storage modules, transportable storage casks, developments in fuel pool storage racks, and integration of wet and dry storage technologies. There was also a presentation by Mikhail Eliakine of IZHORA, the Russian steel fabricator,

Chair's Message
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common understanding of the word. It soon became evident that a common definition of the word was unattainable. Transparency is an end result — the output of many activities — which, in many ways, has a religious nature, much like safeguards. To define safeguards is difficult, yet we all know what it means, both in the domestic and international sense. I jotted down the following thought during the meeting: Transparency is a religion of effective and efficient cooperation regarding the significant knowledge of a state's nuclear activities. This definition is not as clean as one offered by a friend from the Czech Republic: Transparency is what you use with an overhead projector. Cecil Sonnier, chair of the International Safeguards division, intends to prepare a special report on this meeting for publication in a future issue of the *JNMM*. It should be a benchmark article.

As many of you know, the INMM lost two of its staunchest supporters when Jim Jacobs and Leon Green passed away during the Christmas holiday. Included in this issue is a brief note about each man's accomplishments.

The Annual Meeting of the INMM, July 17–20 in Naples, Fla., promises to be exciting. Spread the word about the meeting, especially to those who are new to the world of nuclear materials management.

Should you have comments or questions, please do not hesitate to call me at 505/845-8710.

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describing the company's capabilities to fabricate reactor pressure vessels (as well as other reactor components) and spent fuel shipping casks.

Papers were also presented on the use of the MPC concept in the combined utility/DOE spent fuel management program. This included a description of the basic MPC concepts, the "Green MPC" concept, the impacts of the use of MPCs on utilities and the adaptability of existing canister and cask designs for use in the MPC systems.

The thrust of the current papers focus on improvements in current designs, as contrasted with the addition of new designs that characterized earlier seminars. A number of these improvements appear intended to provide adaptability of the storage systems with the MPC concepts. Having demonstrated the viability of several storage technologies, spent fuel storage system designs are moving toward providing easy integration with the DOE spent fuel management system.

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Committees: N14 and N15

The INMM board of directors received this update on N14 and N15 standards at their board meeting n Chicago, March 1-2.

N14

ANSI N14.1-1990 — *Packaging of Uranium Hexafluoride for Transport*: Randy Reynolds, N14.1 chair, sent correspondence to the writing group with a list of proposed changes and requested input on those changes. The preliminary schedule for N14.1-1995 is:

- Writing group meeting — March 8-9, Knoxville, Tenn.;
- N14 balloting — mid-1994;
- Resolve negative ballots — third quarter CY, 1994; and
- Submit to ANSI for approval and publishing — late 1994.

ISO 7195 — *Packaging of Uranium Hexafluoride for Transport*: This standard was issued on Nov. 1, 1993, and uses much of the N14.1 information; however, it does not include the updated information for N14.1-1990. Metric and English units are used. We will work toward development of an ANSI/ISO standard after a draft of N14.1-1995 is completed.

ANSI N14.2 — *Tiedowns for Transport of Fissile and Radioactive Containers Greater Than One-Ton Truck Transport* (in process): Evaluation and resolution of all comments by the writing group is continuing. It is assumed that the revised draft will require N14 reballoting (probably May 1, 1994).

ANSI N14.5-1987 — *Leakage Tests on Packages for Shipment*: This standard will be balloted for reaffirmation around June 1, 1994. Efforts will continue to develop the draft international standard as an ANSI/ISO N14.5 standard for N14 balloting as a replacement for N14.5. The latest draft, ISO/DIS 12807, was received for balloting, which started Nov. 18, 1993, and ends May 18, 1994. A summary of comments will be provided to N14 members around June 1, 1994.

ANSI N14.6-1993 — *Special Lifting Devices for Shipping Containers Weighing 10,000 Pounds (4,500 kg) or More for Nuclear Materials*: This revised standard was approved by ANSI on June 28, 1993. The new publication is available from ANSI (See address at end of column).

ANSI N14.7 — *Guide to the Design and Use of Shipping Packages for Type-A Quantities of Radioactive Materials*: A plan and schedule are prepared. Plans are for a first draft to be submitted to the writing group by March 1, 1994. Current efforts are directed to complete the writing group. The draft is ready for distribution and will be accomplished after the writing group is completed. A request for additional writing group members was made.

ANSI N14.8 — *Fabricating, Testing and Inspecting Shielded Shipping Casks for Irradiated Reactor Fuel Elements*: This activity will use the peer-panel review to determine standards that should be developed. It is currently not active, but will be activated when documents are received for standards consideration. Completion dates will be set for each document received. Dave Dawson is coordinating, and a recommendation is expected by May 1, 1994.

ANSI N14.10 — *Guide for Liability*: The scope was revised, and the need for the standard is being determined. If needed, a writing group will be formed, and a schedule and draft prepared.

ANSI N14.19-1986 — *Ancillary Features of Irradiated Shipping Casks*: A letter ballot to withdraw this standard was sent to N14 members with a closing date of April 1, 1993. Status of this standard is being evaluated based

Continued on next page

on ballot results. The need for standard is questionable. The inclusion of ISO standard on trunnions will be considered.

ANSI N14.23 — *Design Basis for Resistance to Shock and Vibration of Radioactive Material Packages Greater Than One Ton in Truck Transport*: Work is continuing on preparation of a draft for N14 balloting. The writing group chair feels that the current draft will be suitable for a standard. This decision will be made by the management committee prior to N14 balloting.

ANSI N14.24-1985 (R1993) — *Barge Transport of Radioactive Materials*: This standard was reaffirmed by ANSI, June 28, 1993. Planning for a revised standard is tentatively scheduled for completion by March 1, 1994. A new chair of the writing group is sought and a new scope will be prepared.

ANSI N14.25 — *Tiedowns for Rail Transport for Fissile and Radioactive Material Containers*: Project will start after N14.2 is completed. PINS will be submitted, including a schedule for completion.

ANSI N14.26 — *Guidance on Quality Control Activities as They Relate to the Inspection, Preventive Maintenance and Post-Incident Testing of Packages Used for the Shipment of Radioactive Material*: Work is continuing on preparation of a final draft document. The first draft is complete and the chair is awaiting acceptance of 1985 IAEA regulations by the United States before it is distributed.

ANSI N14.27-1986 (R1993) — *Carrier and Shipper Responsibilities and Emergency Response Procedures for Highway Transportation Accidents Involving Truckload Quantities of Radioactive Material*: Planning started on a new scope and an extensively revised standard. A new writing group chair is needed.

ANSI N14.29-1988 — *For Radioactive Materials – Guide for Writing Operating Manuals for Packaging*: This standard requires extensive revision, and planning to do this has started. The current N14.29-1988 may be balloted for reaffirmation in the event the revision can't be accomplished within three years. This will be decided in the first quarter of 1994. A new writing group chair is needed. An extension to January 1998 was received from ANSI for preparation of a new standard.

ANSI N14.30 — *Design, Fabrication and Maintenance of Semi-Trailers Employed in the Highway Transport of Weight-Concentrated Radioactive Loads*: ANSI approved this standard on Oct. 1, 1992. It is available for sale from ANSI (See address at end of article).

TMD Regulatory Compliance Guide

At the request of J.E. Ratledge, ORNL, a document titled *Guide for Wire Rope Tiedowns on Legal Weight Trucks for Packages Containing Radioactive Materials and Weighing Greater than 5,000 Pounds* is being evaluated for possible development as an ANSI standard. A cursory review indicates that it would be possible to do so. Additional reviews will be made and guidance will be furnished to Ratledge.

Numerical Model Development

Work on development of a numerical model for thermal evaluation of UF6 cylinders is in progress and data is being obtained and analyzed. A draft report when completed will be the basis of an N14 standard. With the transfer of DOE's enrichment operation to the U.S. Enrichment Corp. on Oct. 1, 1993, there was no further activity on this work because of impending funding resolutions.

Standard Matrix

Plans to revise *Standard Matrix for Light-Water Reactor Spent Fuel Transportation* are in progress.

Mixed Waste Standards

Information is being collected on standards that are needed and would be useful for mixed waste packaging and transportation.

N14 Membership

The balloting for eight new N14 members closed on Feb. 16, 1994, and results are being tabulated. Virgil Autry, the representative for the Conference of Radiation Control Program Directors Inc. (CRCPD), resigned and CRCPD is considering a replacement. There are currently 81 members, including 10 alternates, on the N14 committee. There are also 30 people designated "for information only."

N14 Record Retention

A policy for N14 record retention is being drafted, and additional resource material will be incorporated. This policy will be considered for the INMM secretarial policy for both N14 and N15.

ASTM Committee D-10 on Packaging

A liaison between N14 and D-10 was established to avoid conflict and duplication. Robert Towell will serve as the N14 liaison and Robert McGill will serve as the D-10 liaison.

N15

Bruce Moran, Martin Marietta Energy Systems, Oak Ridge, Tenn., is the N15 chair. He was chair of the membership committee. Dean Scott, Westinghouse Hanford Co., Richland, Wash., is the N15 secretary.

A management committee was formed with the following members: Yvonne Ferris, Linda Grady, Bruce Moran, Charles Pietri, Nic Roberts, Dean Scott, Darryl Smith, Michelle Smith and Philip Ting (awaiting confirmation). We could use a few

more volunteers — if you are interested, contact Moran at Martin Marietta Energy Systems, P.O. Box 2009, MS 8206, Oak Ridge, TN 37831-8206, tel: 615/576-8269, fax: 615/574-5169.

The N15 balloting committee was updated and some additional members added. We are trying to add another 10 to 25 at-large members and a few organizational members. Any suggestions for additional members should be sent to John Arendt at 109 Caldwell Dr., Oak Ridge, TN 37830.

Ferris' writing group completed a draft of N15.36-1994 — *Nondestructive Assay Measurement Control and Assurance*. The draft is being balloted until April 1, 1994.

An ad hoc assignment for the management committee is being prepared. It involves a review of 15 standards that were withdrawn, and suggestions for disposition will be requested. The options that will be provided are: new standard, update existing standard, ANSI technical document, potential international standard, priority action or nothing.

John Arendt
INMM/ANSI Nuclear Standards Representative
Oak Ridge Associated Universities
Oak Ridge, Tennessee, U.S.A.

The address for ANSI is:
American National Standards Institute
Attention: Customer Service
11 W. 42nd St.
New York, NY 10036

Divisions: Physical Protection

The quality and quantity of papers received for the 35th Annual Meeting of the INMM, July 17-20, 1994, in Naples, Fla., is encouraging. The division will hold a meeting on Sunday afternoon, July 17, so plan to come early. If you have any suggestions for meeting topics, please contact Division Chair J.D. Williams at 505/845-8766.

The Physical Protection workshop that was tentatively planned for this spring is scheduled for the fall. Details will be available at the annual meeting and in future *JNMM* issues.

J.D. Williams, Chair
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Divisions: Waste Management

More than 150 people attended the INMM Spent Fuel Seminar held Jan. 26-28, 1994, at Loew's L'Enfant Plaza Hotel in Washington, D.C. Topics covered included: spent fuel storage technologies; burnup credit as applied to spent fuel storage and transportation; the multipurpose canister; siting and licensing issues; regulatory and waste management system status; and utility views. Daniel Dreyfus, the new director of the Department of Energy's Office of Civilian Radioactive Waste Management, kicked off the meeting. Richard Stallings, the new U.S. nuclear waste negotiator, also spoke.

A monograph on spent fuel storage was approved by the INMM executive committee. To date, 14 vendors were invited to participate in the preparation of the monograph, with each preparing a chapter on its area of expertise. INMM headquarters will determine the format in which chapters should be submitted and will provide guidelines on their preparation.

The division is putting together six sessions for the annual meeting in July. Sessions on high-level waste, transportation, low-level waste and environmental restoration are scheduled, and a panel discussion on reprocessing is tentative.

The division is looking into holding a Spent Fuel Management Seminar in Japan, sponsored by INMM. The Japanese are very interested in sessions covering burnup credit and multipurpose canisters.

E.R. Johnson, Chair
INMM Waste Management Division
E.R. Johnson Associates, Inc.
Fairfax, Virginia, U.S.A.

Chapters: Pacific Northwest

The chapter elected its 1993–1994 officers:
Chair: Dean D. Scott
Vice-Chair: Scott W. Gority
Secretary-Treasurer: F. Gary Fetterolf
Executive Board: Don E. Six, Dan Nosse, Cindy L. Parnell, Debbie A. Dickman

The chapter recently participated in the local congressional update meeting with Washington Representative Jay Inslee. Regional technical societies met with Inslee to discuss current issues affecting the technical community. The update was an opportunity for technical societies to learn of upcoming initiatives and to provide valuable input to legislators.

Pacific Northwest chapter members also took part in the recent Engineer's Week Celebration, which culminated in a banquet where the chapter was represented.

Upcoming events include the spring chapter technical meeting and the local school science fair. The chapter supports the fair by providing financial support and judges. In addition, local INMM members are working with Sandia Laboratory in Albuquerque, N.M., to coordinate an integrated workshop on long-term storage facilities to be held later this year.

Debbie Dickman
Pacific Northwest Laboratory
Richland, Washington, U.S.A.

Vienna

Retiring International Atomic Energy Association (IAEA) Safeguards Director Ray Parsick addressed the December luncheon meeting of the INMM Vienna Chapter. He spoke on catastrophic accidents and safeguards. He said that the world is demanding a stronger safeguards system, and this will involve a great deal of work. Subsequent comments from the audience as to the nature of this new safeguards system were lively, attesting to the interest which the address provoked.

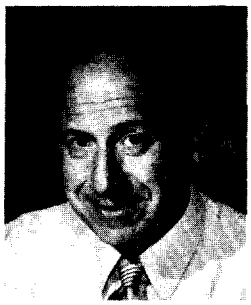
While Parsick left IAEA, he did not leave the world of work. He returned at the beginning of February to Brookhaven National Laboratory.

Ed Kerr
IAEA (retired)
Vienna, Austria

Japan

More than 140 people attended the 14th Annual Meeting of the Japan Chapter, Nov 9-10. K. Ikawa of the Japan Atomic Energy Research Institute served as the program chair, and Japan Chapter Chair T. Haginoya spoke on the situation of nonproliferation and the role of international safeguards. Session topics included reprocessing safeguards and LEU facility safeguards.

Member News



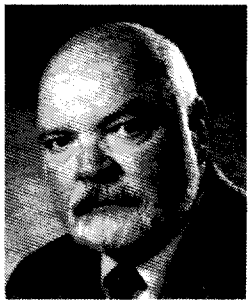
Leon Green, the previous head of the International Safeguards Project Office (ISPO) at Brookhaven National Laboratory, died on Dec. 8, 1993, after a brief illness. He was 70.

Green came to Brookhaven on Oct. 24, 1956, as a guest engineer. By 1966, he was leader of the evaluation and technical assistance group, which made engineering and economic evaluations of new power-reactor concepts.

From 1966-68 he took a leave of absence to work in Vienna in the economic section of the International Atomic Energy Agency's (IAEA) department of nuclear power and reactors. Returning to Brookhaven in 1968, he joined the newly formed Technical Support Organization (TSO), established by Herbert Kouts and William Higinbotham. In 1977, he was named an associate chemical engineer and eventually chemical engineer. Also that year, the ISPO was established within the Department of Nuclear Energy (DNE), and, one year later, Green was appointed its second head.

In July 1989, Green received the INMM Distinguished Service Award from the INMM. The IAEA recognized him with a signed plaque for 13 years of support to IAEA safeguards.

In December 1991, Green stepped down from his leadership of ISPO but continued to advise the office and contribute to other activities within DNE, which became the department of advanced technology in 1993.



James "Jake" Jacobs passed away on Dec. 26, 1993, at the age of 59. He was one of few individuals in the United States who combined the expertise in nuclear security systems technology with a thorough knowledge of relevant domestic, foreign and national security problems and policies.

Jacobs joined Sandia National Laboratories in 1959. In 1964, he was one of several Sandia employees who took a leave of absence to help Bellcomm Inc. work on systems development for the Apollo project. In 1969, he became supervisor of the advanced components development division. In 1977, he was promoted to manager of the advanced systems department. He was named director of the nuclear security systems directorate in August 1988 and transferred to become director of facilities program management center in 1991.

In 1986, he received the INMM Distinguished Service Award. In 1991 he was named a Fellow of the INMM. In early December 1993, the U.S. Army honored Jacobs by presenting him the Army's Outstanding Civilian Service Medal.

Pacific-Sierra Joins INMM

A new sustaining corporation was approved. The INMM welcomes Pacific–Sierra Research Corp., located in Arlington, Va.

Corrections

In the January issue of *JNMM*, John Bartlett was incorrectly associated with Analytic Sciences Corp. He works for E.R. Johnson Associates Inc., Fairfax, Va. Also in the January issue, John L. Jaech's affiliation was listed incorrectly. He works for Safestat Inc.

International News

The European Safeguards Research and Development Association (ESARDA) is celebrating its 25th anniversary this year.Its partners, as of May 1, 1993, are:

- The European Atomic Energy Community;
- The Kernforschungszentrum Karlsruhe (KfK), Germany;
- The Centre d'Etude de l'Energie Nucléaire – Studiecentrum voor Kernenergie (CEN/SCK), Belgium;
- The Ente per le Nuove Tecnologie, l'Energia e l'Ambiente (ENEA), Italy;
- The Stichting Energieonderzoek Centrum Nederland (ECN), Netherlands;
- The Atomic Energy Authority (AEA), United Kingdom;
- The Commissariat à l'Energie Atomique (CEA), France;
- The British Nuclear Fuels plc (BNFL), United Kingdom;
- The Forschungszentrum Jülich GmbH (KFA), Germany; and
- The Centro de Investigaciones Energéticas Medioambientales y Tecnologicas (CIEMAT), Spain.

Low-Level Radioactive Waste Treatment and Disposal

Pierre Saverot
NUSYS
Paris, France

Editors Note: *This paper represents part one of a two-part series discussing various aspects of low-level radioactive-waste (LLW) disposal, written by Pierre Saverot, chairman of the Low-Level Waste Disposal Committee of the INMM Waste Management Division. Saverot is associated with the French consulting firm NUSYS, which is involved in technical and management consulting on the back-end of the fuel cycle management matters, among its other activities, and has been active in nuclear engineering for more than 15 years.*

The second paper of this series will deal with boundary conditions for release pathways and safety analyses for a LLW disposal facility and will be published in a subsequent issue of the JNMM.

Abstract

Major national regulatory guidelines applicable to low-level radioactive waste (LLRW) disposal are implemented to protect the public health and the environment from the radioactive substances during the lifetime of the disposal facility. These guidelines translate into waste acceptance criteria i.e., physical stabilization of the waste form, containment or immobilization of the radionuclides in the waste. A waste acceptance process is enforced on LLRW generators to determine the suitability of the waste form in meeting the stabilization and immobilization criteria. This paper discusses the waste characteristics and the waste acceptance criteria for LLRW disposal. Although the figures and numbers (such as radionuclide concentrations) quoted are derived from the French practice, it is felt that the issues considered and discussed are applicable to any comprehensive cradle-to-grave low-level waste management system.

Introduction

Radioactive waste is defined by the International Atomic Energy Agency [2] as "Any material that contains or is contaminated with radionuclides at concentrations or radioactivity levels greater than the 'exempt quantities' established by the regulatory body and for which no use is foreseen."

Various classification systems are used to categorize radioactive wastes. Materials considered as waste are those declared as having no immediate technical, economical and/or political use and are, therefore, disposed of for a long period of time. Intermediate stored products separated during the reprocessing operations, such as uranium and plutonium, which are or will be reused, are not considered wastes. Classification systems have been based on radionuclides content, origin of the waste, type of radiation, half-life of the nuclides, hazardous lifetime of the waste, radiotoxicity, specific activity (in terms of mass and volume) and dose rate. Each of these classification systems has its advantages, and thus most countries have developed their own classification systems designed to suit the conditions and regulations prevailing in that country.

The terms *low-level*, *intermediate* (or *medium*)-*level* and *high-level* radioactive wastes are widely used to describe different concentrations of radioactive materials in wastes. Unfortunately, these terms do not have quantitative definitions, and in this way confusion arises when reference is made simply to high-, intermediate- or low-level wastes undefined by concentration of radioactivity level. In various individual countries, different considerations have determined the systems of classification used for radioactive waste. These considerations include environmental limitations for acceptance of waste and existing operational situations in terms of waste type and waste treatment systems. The different systems in turn lead to different regulations and make it difficult for communication between countries on waste management topics.

As an example, the classification of radioactive wastes in the United States is as follows:

(1) High-level waste (HLW): "the highly radioactive material resulting from the reprocessing of spent fuel, including liquid waste produced directly in reprocessing, and

any solid material derived from such liquid waste, that contains fission products in sufficient concentration, and other highly radioactive materials that the NRC, consistent with existing law, determines, by rule, to require permanent isolation." In a once-through cycle, the spent nuclear fuel is considered to be HLW.

(2) Transuranic waste (TRU): the NRC defines TRU waste as waste containing alpha-emitting isotopes with atomic number greater than 92, with half-lives greater than five years and concentration greater than 3.7×10^6 Bq/kg (100 nCi/g waste). The U.S. Department of Energy (DOE) and the U.S. Environmental Protection Agency (EPA) use a slightly different definition, with the same specific activity but half-lives greater than 20 years.

(3) Low-level waste (LLW): LLW is defined "as not HLW, spent nuclear fuel, transuranic waste or byproduct material as defined in section 11e-2 of the Atomic Energy Act of 1954; and material the NRC, consistent with existing law, classifies as LLW."

(4) Uranium mill tailings are also considered radioactive wastes.

(5) Another category of waste not regulated by the NRC but only by individual states is the naturally occurring or accelerator-produced radioactive material (NARM), which classifies as LLW in terms of disposal.

In France, radioactive wastes are classified in three categories:

Category A: low- and intermediate-activity waste containing short-lived radionuclides (less than 30 years half-life) and containing only traces of long-lived radionuclides (a 3.7 GBq/t). These wastes are sent to a surface repository, with a post-closure control period of not more than 300 years expected.

Category B: intermediate radioactive waste containing long-lived radionuclides (greater than 30 years half-life), mostly alpha-emitting radionuclides, with no significant heat output, categorized as low-heat TRU waste, where the TRU breakpoint is 100 nCi/g maximum content with an average waste loading in a storage site not exceeding 10 nCi/g.

Category C: high-level radioactive waste; waste containing fission products resulting from spent fuel reprocessing, solidified by vitrification and producing, at the time of its

Table 1: Classification of radioactive wastes

FRANCE	UNITED KINGDOM	USA	JAPAN	ITALY
High Level Waste C Vitrified fission products with heat generation.	High Level Waste Heat generating.	High Level Waste Above low.	High Level Waste Primary extracted liquid containing fission products from spent fuel reprocessing and its vitrified product.	High Level Waste (III Category) Waste requiring thousands of years to decay to a radioactive concentration of some hundreds of Bq/g (waste arising from reprocessing facilities and wastes containing alpha and neutron emitters).
Intermediate Level Waste B >3.7 GBq/t half life >30 y mainly alpha emitters no appreciable heat generation.	Intermediate Level Waste Above LLW but for which heat generation does not have to be considered.			Low Medium Level Waste (II Category) Wastes requiring a period varying from a few decades to a few centuries to decay active concentration of some hundreds of Bq/g (mainly arising in NPP operation or in some industrial and research activity).
Low Level Waste A Half life < 30 years alpha <3.7 GBq/t	Low Level Waste For alpha emitters <4 GBq/t For beta/gamma emitters <12 GBq/t	Low Level Waste <3.7 GBq/t for alpha nuclides with half lives >5 years <130 GBq/t for Cm-242 <740 GBq/t for Pu-241 Sub-classes A, B and C	Low Level Waste All wastes not defined as high level waste.	Low Level Waste (I Category) Wastes requiring a maximum period of a few years to decay to a radioactivity concentration depending on nuclide radiotoxicity. The Italian law establishes the following limits: 0.37 Bq/g for very high radiotoxicity (*) 3.7 Bq/g for high radiotoxicity 37 Bq/g for moderate radiotoxicity. 370 Bq/g for low radiotoxicity (**) This category of wastes mainly produced in biomedical and research activity. (*) Like Pu, Am, Cf, Cm, etc. (**) Like U-235, Cs-135, etc.

fabrication, a large heat output.

Wastes of category B and C are currently being stored prior to geological disposal.

In order to emphasize the need for caution in using terms like low, intermediate and high in any specific context, the waste classification definitions for a number of countries are outlined in Table 1 (below).

Major regulatory guidelines applicable to LLW disposal define two basic performance criteria: (1) protection of public health and of the environment from radioactive substances during the three periods (operational, institutional control and unrestricted site access) that make up the lifetime of the disposal facility, and (2) limitation of the time required for the surveillance of the disposal facility to 300 years (500 years in the United States).

Based on these objectives, the performance objectives translate into three primary waste acceptance criteria. The first is to physically stabilize the waste form. The second is to contain or immobilize the radionuclides in the waste. The third is to limit the specific activity of short-lived (0.5- to 6-year half-life), intermediate-lived (6- to 30-year half-life), and long-lived (greater than 30-year half-life) radionuclides in the waste.

The LLW generators must enforce a waste acceptance process to make a determination about the suitability of the waste form in meeting the first two criteria defined above (stabilization and immobilization). The third criteria results in the development of maximum values for specific activities of the waste. The limit for each radionuclide is different and depends on the radiotoxicity of each radionuclide; results of pathways analyses for the free access period imply the necessity of limiting the calculated maximum specific activity of long-lived alpha emitters to less than 0.1 Ci/ton for individual packages and to less than 0.01 Ci/ton for individual disposal units.

Waste Characteristics

Examples of waste types intended for disposal include ion-exchange resins stabilized and immobilized in organic polymer and packaged in concrete overpacks; filters stabilized and immobilized in cement and packaged in concrete overpacks; concentrates or sludge stabilized and immobilized in bitumen and packaged in metal drums; and miscellaneous solid wastes stabilized and immobilized in cement and packaged in drums, concrete overpacks or metal boxes. Conditions for acceptance of solid immobilized waste for near surface disposal include two waste types, homogeneous and heterogeneous. Homogeneous waste is evenly distributed throughout the solidification matrix, such as ion-exchange resins immobilized in polymer. Heterogeneous waste is in discrete pieces that are "blocked" inside a container by the solidification material, such as dry active waste that is cemented.

Radioactive Containment

The radioactive containment performance of a homogeneous waste form is measured by its leach rate, which is determined by leach tests performed over a minimum period of one year in accordance with standard test procedures. The average leach rate objectives are as follows:

- 6×10^{-10} m/s for each beta-gamma emitting radionuclide important for safety, except tritium, when the specific activity of the waste for this radionuclide is between the immobilization threshold and 37 MBq/kg.
- 1.2×10^{-10} m/s for each beta-gamma-emitting radionuclide important for safety, except tritium, when the specific activity of the waste for this radionuclide is between 37 and 370 MBq/kg (2.4×10^{-11} m/s when the specific activity is greater than 370 MBq/kg).
- 1.2×10^{-12} m/s for each alpha-emitting radionuclide important for safety.

For purposes of safety analyses, the container of the homogeneous waste form is ignored. A metal drum will perish. Similarly, a concrete overpack adds stability and provides radionuclide immobilization, but it is not taken into account. It is the homogeneous waste form itself that accomplishes this. If the container is stripped away, a surface consisting of a mixture of the solidification matrix plus the homogeneous waste is left. For this type of surface, a long-term leach test (450-plus days) is the principal test for immobilization. The fraction of radioactivity released annually (FAL) is the principal figure of merit by which to judge the ability of the waste form to immobilize and contain radionuclides. Examples of homogeneous waste forms include ion-exchange resins, evaporator concentrates and coprecipitation sludges.

For heterogeneous waste forms, radioactive containment performance is expressed in terms of the fraction of activity leached annually, taking into account both the waste form and the encapsulation material. Regulations establish the same objectives for heterogeneous waste as for homogeneous waste but require a case-by-case assessment of the waste forms. In addition, the long-term stability of the container, of its closure system and of any encapsulation material between the container and the waste form must be evaluated. If the container is stripped away from the heterogeneous waste form, the encapsulation layer around the waste/grout mixture remains. A diffusion test that measures the rate of radionuclide migration across the encapsulation layer is the principal figure of merit for this waste form. The diffusion test of the encapsulation material lasts for one year. Tritium and Cesium-137 are the two isotopes used in the test. Examples of heterogeneous waste forms include water purification filters, ventilation filters, gloves, rags and other dry active waste.

Compressive Strength

All waste packages must be capable of supporting a load of 0.35 MPa (50 psi) with less than 3% vertical deformation.

Resistance to Thermal Cycles
The variation of mechanical resistance after thermal cycling must be less than 20% for both waste forms. If the homogeneous waste form shows obvious or measured defects after the thermal test, additional leach tests must be performed.

Resistance to Beta/Gamma Radiation
Homogeneous and heterogeneous waste forms with a contact dose rate greater than 50 rad/h, irradiated with a dose from 2×10^4 to 1×10^7 rad, must show a variation in mechanical resistance of less than 20%. For both waste types, the characteristics of the waste must be such that gas generated by radiolysis does not affect the integrity of the container.

Type and Activity of Radionuclides
With respect to the chemical nature of the waste, the generator must identify significant quantities of complexing agents, particularly chelating agents, and the waste must be treated to reduce the agents or inhibit their complexing properties. The waste may not contain toxic chemical or biological substances or pyrophoric substances that can create a spontaneous exothermic reaction. Waste may not contain organic liquids or free-standing aqueous liquids. The amount of water released under compression of 0.35 MPa may not exceed 1% by volume. In addition, the waste must be in a form compatible with the immobilization material (cement, grout, bitumen or organic polymer). In particular, sodium, aluminum and magnesium may cause problems because of incompatibility with common immobilizing materials. For LLW that contains fissile isotopes, nuclear criticality safety must be respected and, of particular concern, is uranium with an enrichment of greater than 1%: the amount of fissile material must not exceed 0.1 grams per kilogram of waste form divided by the density of the waste form.

With respect to the type and activity of radionuclides, the generator must determine these by using one or several methods, such as direct determination, detection of certain radionuclides and correlation with others, and standard spectra. Activity limits are established for principal radionuclides, with a ceiling for all short-lived beta/gamma radionuclides of 1 Ci/t, and a ceiling for all long-lived alpha-emitting radionuclides of 0.005 Ci/t (the mass used is the immobilized waste form plus the container plus any encapsulation material). Waste packages containing high concentrations of long-lived beta/gamma emitters, waste with more than 0.1 Ci/t alpha or waste with more than 2 Ci/t of tritium are subject to special acceptance based on the results of pathways analysis.

Minimum Requirements for a Characterization Program

Characterization of a generator's waste stream consists of all the actions taken to demonstrate that safety regulations and technical specifications have been satisfied by both the full-scale process for producing the waste form, and the waste form itself. The characterization program is set up in such a way as to assure that the process and its product (the waste form) continue to meet applicable evaluation criteria in the future. The radioactive characteristics (isotopes present, activity level and chemical composition) of the waste must be identified at the beginning of the characterization program. In general, the minimum characterization criteria are divided into four categories: physical properties, mechanical properties, containment or radionuclide retention capacity, stability and alterability. This last category deals with the ability of the waste form to maintain its radionuclide retention capacity over long periods of time.

Physical Properties
Physical properties of the stabilization or immobilization material:

- density, porosity, permeability to water, permeability to gases;
- thermal conductivity, change of state temperature; and
- solubility in water, exudation of water under compressive stress, shrinkage and curing weight loss.

Physical properties of the stabilization or immobilization material plus waste:

- density, porosity, permeability to water, permeability to gases;
- homogeneity, compatibility of the waste with the grout, cohesion;
- change of state temperature; and
- percentage of water incorporated, exudation of water under compressive stress, shrinkage and curing.

Physical properties of material making up the container:

- density, porosity, permeability to water, permeability to gases;
- thermal conductivity, change of state temperature; and
- solubility in water, exudation of water under compressive stress, shrinkage and curing weight loss,

Physical properties of the package (waste plus grout plus container):

- amount of voids in the container (minimize), and
- characteristics of lid and seal plug.

Mechanical Properties
Mechanical properties of the stabilization or immobilization material:

- tensile strength, compressive strength, hardness.

Mechanical properties of stabilization or immobilization material plus waste:

- tensile strength, compressive strength, hardness.

Mechanical properties of the package (waste plus grout plus container):

- behavior under load, impact strength.

Radionuclide Retention Capacity
Retention capacity of the immobilization material:

- diffusion of radionuclides in aqueous medium;

- diffusion of tritium under standard atmospheric conditions; and
- radionuclide fixation and retention capability.

Retention capacity of the immobilization material plus waste:

- leaching of radionuclides in aqueous medium;
- gas release under standard atmospheric conditions; and
- radionuclide fixation and retention capability.

Retention capacity of the container:

- diffusion of radionuclides in aqueous medium, and
- diffusion of tritium under standard atmospheric conditions.

Retention capacity of the package (waste plus grout plus container):

- water-tightness of the package sealing device, and
- gas release under standard atmospheric conditions.

Stability and Alterability — Maintenance of the Retention Capacity of the Package

Long-term retention capability of the stabilization or immobilization material:

- behavior under temperature cycling, under beta/gamma radiation, and
- sensitivity of the grout to water contact (swelling, weight change, surface texture change, changes in the cohesion between the grout and the waste).

Long-term retention capability of the immobilization material plus waste:

- behavior under temperature cycling, under beta/gamma radiation;
- sensitivity of the grout to water contact;
- sensitivity to elevated temperatures; and
- resistance to the action of microorganisms.

Long-term retention capacity of the material making up the container:

- corrosion resistance in wet medium (for metal drums);
- porosity, degree of gas-tightness; and
- behavior under beta/gamma radiation.

Long-term retention capacity of the package (waste plus grout plus container):

- behavior under temperature cycling, and
- fire behavior.

Note that the term *grout* refers to either bitumen, cement or organic polymer. The term *grout plus waste* refers to the homogeneous or heterogeneous mixture of waste plus grout material that together form the stabilization and immobilization matrix. Encapsulation material is the same as grout material; however, occasionally encapsulation material is specifically called out to emphasize that it is present and being tested.

Table 2: Acceptance limits for long-lived alpha emitters

RADIONUCLIDES	IMMOBILIZATION THRESHOLD		ACCEPTANCE LIMIT	
	MBq/kg	Ci/t	MBq/kg	Ci/t
^{226}Ra	0.037	0.001	3.7	0.1
^{232}Th	0.037	0.001	1.1	0.03
Total Radionuclides	0.19	0.005	3.7	0.1

Waste Acceptance Criteria

A primary design goal of a LLW disposal facility is to keep water away from the waste, because water transport is the principal mechanism for radionuclide migration. Collapsing of voids within waste packages can lead to collapse and subsidence of portions of the final disposal cap. Subsidence of the disposal cap invites rainwater pooling and increased rainwater infiltration into the disposal system. Furthermore, if waste packages crack under load, the infiltrated water may leach out radionuclides from the waste. Void reduction of dry active waste and contaminated scrap can be achieved by such processes as shredding, compacting and incineration; stability is added to the resulting waste products by cement grouting of the waste. The principal test for stabilization is resistance of the final waste package to a load of 0.35 MPa (50 psi) with little or no deformation (less than 3%) and with little or no liquid release under compression.

The second waste acceptance criterion is to contain or immobilize the radionuclides present in the waste. For homogeneous waste, one method for achieving immobilization is the development of solidification matrices (cement, bitumen and polymer) that will fix the radionuclides in the homogeneous mixture and resist leaching. *Heterogeneous waste* must be encapsulated, and the development of encapsulation materials that resist radionuclide diffusion is an important means of achieving immobilization. In France, LLW is classified as being above or below an immobilization threshold in Curies per ton.

The immobilization threshold for each radionuclide is chosen as a result of pathways analysis; it is a risk-based concept in which engineering effort is expended where it will most significantly reduce potential exposure and dose to humans: waste with a specific activity greater than the immobilization threshold determined for each radionuclide must be immobilized while waste that is below the immobilization threshold may be simply grouted and stabilized.

The third waste acceptance criterion is a limitation on the specific activity of short-lived, intermediate-lived and long-

Table 3: Immobilization threshohold for short- and intermediate-lived alpha emitters

RADIONUCLIDES	IMMOBILIZATION THRESHOLD	
	MBq/Kg	Ci/t
Any Individual Radionuclide	3.7	0.1
Total Radionuclides	37.0	1.0

lived radionuclides in the waste. This is the *upper activity limit* for waste acceptance: waste that is above this limit is not accepted for disposal. Tables 2, 3, and 4 provide examples of acceptance limits. These limits are set on specific activities such that detailed pathways analyses for all periods in the life of the facility (operating, institutional control and unrestricted site access periods) demonstrate that for both normal and accidental conditions the radiological impacts of the facility are acceptable.

For beta/gamma emitters with half-lives of less than 30 years, the acceptance limit for the specific activity at the time of acceptance at the disposal facility for total radionuclides varies as a function of the acceptance limits for individual radionuclides contained in the waste packages. If one radionuclide is above the acceptance limit, then the entire waste package is considered to be above the acceptance limit. If there are more than 10 radionuclides present, then the sum of fractions (i.e., the actual specific activity of the radionuclide divided by its acceptance limit) for the waste package must be less than 10.

Whenever waste contains significant amounts of beta/gamma emitters not specifically listed in the tables, the individual radionuclide limits that will be applied to this waste are derived from a special safety study submitted to the safety authorities for approval. An example would be a waste package containing significant quantities of Ni-63, which would be subject to a special review.

Radiological Inventory

The radiological inventory of a LLW disposal facility is an estimate of the total quantities and types of radionuclides considered important by the safety authorities that are expected to be present in all waste packages requiring disposal (H-3, C-14, Co-60, Ni-63, Sr-90, Nb-64, Cs-137, Np-237, U-238, Pu-239, Am-241, ...). It is used in the safety analyses of the site and may be modified during the iterative process involved in the pathways analyses. If for a given radiological inventory the radiological impacts are considered to be unacceptable, then the inventory is reduced for some or several radionuclides. The radiological inventory that is ultimately approved as a result of pathways analysis showing that its impacts are acceptable becomes the radiological capacity of the disposal facility, which represents a specification for the maximum allowable quantities of individual radionuclides considered important for safety for that disposal facility.

Table 4: Examples of acceptance limits for intermediate-lived beta/gamma emitters

INTERMEDIATE-LIVED BETA-GAMMA EMITTERS	IMMOBILIZATION THRESHOLD		ACCEPTANCE LIMIT	
	MBq/kg	Ci/t	MBq/kg	Ci/t
^{3}H (12.3 year)	7.4	0.2	$7.4\ 10^{1}$	2.0
^{22}Na (2.58 year)	20.0	0.5	$2.0\ 10^{5}$	$5.4\ 10^{3}$
^{55}Fe (2.7 year)	37.0	1.0	$3.6\ 10^{6}$	$8.1\ 10^{4}$
^{60}Co (5.27 year)	3.7	0.1	$4.8\ 10^{4}$	$1.3\ 10^{3}$
^{90}Sr (28.2 year)	3.7	0.1	$7.4\ 10^{2}$	$2.0\ 10^{1}$
^{106}Ru (1.0 year)	9.0	0.25	$8.8\ 10^{4}$	$2.4\ 10^{3}$
^{102}Rh (2.9 year)	20.0	0.5	$2.0\ 10^{5}$	$5.4\ 10^{3}$
^{137}Cs (30.2 year)	3.7	0.1	$4.8\ 10^{3}$	$1.3\ 10^{2}$
^{210}Pb (22.3 year)	0.04	0.001	$4.0\ 10^{1}$	1.1
^{228}Ra (5.8 year)	0.1	0.003	100	2.7

Determination of Errors Present in a Mass Spectrometric Evaluation of Uranium Isotopes*

Andrew White
Argonne National Laboratory
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Abstract

In this article the magnitude of the error in a mass spectrometric evaluation of uranium content at the Argonne National Laboratory-West (ANL-W) analytical laboratory is determined. The quantities being evaluated are the elemental uranium fraction and the isotopic fractions of each of the uranium isotopes. The basic uncertainties in the operations of each of these three phases are identified. The uncertainties in the different phases are propagated using both linear and Monte Carlo techniques and result in long-term systematic, short-term systematic, and random errors, respectively. Underlying uncerainties include those resulting from weighing, uncertainties in standard material composition, and uncertainties introduced by use of the mass spectrometer from atom ratio measurements.

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Introduction

Mass spectrometry is used at ANL-W to determine the uranium isotopic fractions of a sample. These fractions are needed for chemical research, process control and for material accountability. For all of these applications, a knowledge of the error in the measurement is desirable. For the case of material accountability it is required.

The typical uranium sample that is processed in the analytical laboratory is taken from a fuel pin. These pins contain approximately 70% U-235, 29% U-238, 0.6% U-234, and 0.2% U-236. This composition is used in the error propagation calculations below.

There are three phases in the evaluation of the uranium content. Preparation of a standard is performed approximately once every 20 years. The second phase, mass spectrometer and spike calibration, is performed approximately once a year. The third phase, the sample evaluation, can be performed several times per day. The following sections give a brief description of each of the phases. In each section, the analytical expressions associated with the step are given.

The error propagation analysis is based on the standard equation for the linear propagation of errors:

$$\sigma_{tot}^2 = \sum_i \left(\frac{\partial}{\partial X_i} f(X) \right)^2 \left(\sigma_{X_i} \right)^2 + \sum_i \sum_{j,j \neq i} \frac{\partial}{\partial X_j} f(X) \frac{\partial}{\partial X_j} f(X) \sigma_{X_i, X_j}$$

(Equation 1)

where

σ_{tot} is the total standard deviation,

X is the vector of nominal parameters,

f(X) is the function which relates the response to the input variables,

σ_{X_i} is the standard deviation for parameter i, and

σ_{X_i, X_j} is the covariance between parameters X_i and X_j.

Because the equations are nonlinear, it is not possible to divide the expressions into easy-to-handle, smaller expressions. The complexity of the expressions also necessitates the use of numerical rather than analytical differentiation.

Standard Preparation

As mentioned above, the frequency of the standard preparation is once every 20 to 30 years. There are two standards that must be prepared. The first, certified reference material CRM U500, contains uranium with a U-235 to U-238 atom ratio of 0.9997 ± 0.1% and is used to calibrate the instrument. The second, NBS960, contains natural uranium metal (99.975% pure, with an isotopic composition of 99.2749 ± 0.0003 U-238, 0.7197 ± 0.0003 U-235, and 0.00544 ± 0.00002 U-234, according to ref. 2, and is used to calibrate the U-233 spike.

The only preparation required for the CRM U500 standard is its dissolution and dilution to the proper working specifications for the mass spectrometer. The dissolution and dilution result in no change to the U-235 to U-238 atom ratio. Because this ratio is the only quantity of concern, the manner in which the material is diluted does not contribute to the error.

The NBS960 standard solution is prepared by dissolving the material in an acid solution. Unlike the CRM U500 standard, the exact quantity of NBS960 standard in the solution is needed for this step. Therefore, accurate measurement of the amount of material in the standard and the amount of solution added is required. The formula for the resulting standard concentration is:

$$C_{sol_u^{NBS}} = \frac{\left(M_{met}^{NBS} - M_{met_t}^{NBS} \right) \times Pur^{NBS}}{M_{dil}^{NBS} - M_{dil_t}^{NBS}}$$

(Equation 2)

where

$C_{sol_u}^{NBS}$ is the concentration of uranium in the NBS solution,

M_{met}^{NBS} is the mass of metal to be dissolved in the standard solution plus the mass of the container,

$M_{met_t}^{NBS}$ is the tare mass of the container which held the metal,

M_{dil}^{NBS} is the total mass of the dissolved standard and solution,

$M_{dil_t}^{NBS}$ is the tare weight of the dilution container, and

Pur^{NBS} is the weight fraction of uranium in the NBS (now NIST) standard.

Mass Spectrometer and Spike Calibration

The purpose of performing the mass spectrometer calibration is to obtain the bias correction factor for the instrument. The purpose of performing the spike calibration is to determine as accurately as possible the composition of the spike solution. The two calibrations are discussed together because they are performed at the same time. There are four steps in these calibrations: the determination of the mass spectrometer discrimination bias correction, the determination of the weight fractions of U-235 and U-238 in NBS960, the determination of the weight fractions of the uranium isotopes in the U-233 spike, and the determination of the uranium concentration in the U-233 spike. The first of these steps is associated with the mass spectrometer calibration and the final three are associated with the spike calibration. The discrimination bias impacts all measurements of the mass spectrometer and therefore must be determined first. The weight fractions of U-235 and U-238 in the NBS960 and the weight fraction of the uranium isotopes in the spike are needed in the determination of the concentration of uranium in the spike solution.

Mass Spectrometer Discrimination Bias Correction

This bias correction accounts for a particular instrument bias in the determination of isotopic ratios. The equation used to determine the bias is:

$$B = \frac{1}{3} \left(\frac{R85NBS}{R85ANL} - 1 \right)$$

(Equation 3)

where

B is the mass spectrometer discrimination bias correction (per amu),

R85NBS is the atom ratio of U-238 to U-235 as certified by NBS, and,

R85ANL is the atom ratio of U-238 to U-235 as measured by ANL-W.

The factor of ⅓ in the above equation is included because there is a 3 amu difference between U-235 and U-238 and

the bias is expressed in units of amu^{-1} or (atomic mass units)$^{-1}$.

NBS960 Isotopic Weight Fractions

The isotopic weight fraction of U-238 in the NBS960 standard is determined by evaluating a sample of the standard in the mass spectrometer. The weight fraction is computed using:

$$W_{U238}^{NBS} = \frac{1}{1 + R48\left(\frac{1}{1+4B}\right)\left(\frac{234}{238}\right) + R58\left(\frac{1}{1+3B}\right)\left(\frac{235}{238}\right)}$$

(Equation 4)

where

- B is the mass spectrometric discrimination bias (per amu),
- R48 is the atom ratio of U-234 to U-238,
- R58 is the atom ratio of U-235 to U-238,
- W_{U238}^{NBS} is the weight fraction of U-238 in the NMS 960 standard, and
- $\frac{23X}{238}$ is the atomic mass ratio of U-23X to U-238.

U-233 Spike Isotopic Weight Fractions

The weight fraction of U-233 in the spike is determined by placing a sample of the spike solution in the mass spectrometer. The equation expressing the weight fraction of U-233 in the spike is:

$$W_{U233}^{Spike} = \frac{1}{1 + R43(1+B)\frac{234}{233} + R53(1+2B)\frac{235}{233} + R63(1+3B)\frac{236}{233} + R83(1+5B)\frac{238}{233}}$$

(Equation 5)

where

- R43 is the atom of U-234 to U-233,
- R53 is the atom ratio of U-235 to U-233,
- R63 is the atom ratio of U-236 to U-233,
- R83 is the atom ratio of U-238 to U-233, and
- W_{U233}^{Spike} is the weight fraction of U-233 is the spike.

The weight fraction of the other isotopes in the spike can be expressed as

$$W_{U23X}^{Spike} = \frac{RX3(1+(X-3)B)23X}{233 + R43(1+B)234 + R53(1+2B)235 + R63(1+3B)236 + R83(1+5B)238}$$

(Equation 6)

where

- W_{U23X}^{Spike} is the weight fraction of U23X in the spike.

Uranium Concentration in the U-233 Spike

The concentration of uranium in the U-233 spike solution is determined by adding a known quantity of NBS960 standard solution to a known quantity of spike solution. Because the weight fractions of the isotopes in each of the mixture components is known, and the concentration of uranium in the NBS960 is known, it is possible to determine the concentration of uranium in the spike using:

$$C_{sol_u}^{Spike} = \frac{C_{sol_u}^{NBS} M_{cmix}^{NBS}}{M_{cmix}^{Spike}} \left[\frac{W_{U238}^{NBS} - W_{U238}^{cmix}}{W_{U238}^{cmix} - W_{U238}^{Spike}} \right]$$

(Equation 7)

where

- W_{U238}^{cmix} is the weight fraction of U-238 in the calibration mixture,
- $C_{sol_u}^{NBS}$ is the concentration of uranium in the NBS solution,
- M_{cmix}^{NBS} is the mass of the NBS 90 solution in the calibration mixture,
- $C_{sol_u}^{Spike}$ is the concentration of uranium in the spike solution,
- M_{cmix}^{Spike} is the mass of the spike in the calibration mixture,

Continued on next page

W_{U238}^{NBS} is the weight fraction of isotope i in NBS960, and

W_{U238}^{Spike} is the weight fraction of U-238 in the spike solution.

The weight fractions for the different isotopes in the mixture can be calculated once the atom ratios of the isotopes are measured.

Substituting for the various quantities, the following expression is obtained:

$$C_{sol_u}^{Spike} = \frac{M_{sam}^{NBS} C_{sol_u}^{NBS}}{M_{sam}^{Spike}} \left[\frac{\dfrac{R38'\left(\frac{233}{1+5B}\right)+R48'\left(\frac{234}{1+4B}\right)+R58'\left(\frac{235}{1+3B}\right)+R68'\left(\frac{236}{1+2B}\right)+238}{\left(238+\frac{234R48}{1+4B}+\frac{235R58}{1+3B}\right)} - 1}{1-\dfrac{R83(1+5B)\left\{R38'\left(\frac{233}{1+5B}\right)+R48'\left(\frac{234}{1+4B}\right)+R58'\left(\frac{235}{1+3B}\right)+R68'\left(\frac{236}{1+2B}\right)+238\right\}}{\left[238+R43(1+B)234+R53(1+2B)235+R63(1+3B)236+R83(1+5B)238\right]}} \right]$$

(Equation 8)

where

R38' is the atom ratio of U-233 to U-238 in the 1:1 spike to standard calibration mixture,

R48' is the atom ratio of U-234 to U-238 in the 1:1 spike to standard calibration mixture,

R58' is the atom ratio of U-235 to U-238 in the 1:1 spike to standard calibration mixture, and

R68' is the atom ratio of U-236 to U-238 in the 1:1 spike to standard calibration mixture.

Sample Evaluation

The goal in the evaluation of a sample is to determine its isotopic and elemental composition. The procedure and equations used to determine this are given below.

Procedure

The sample evaluation requires sample cleaning, sample dissolution, spike and sample combination and chemistry, and mass spectrometric evaluation. The procedures for each of these steps are given below.

Sample Cleaning Procedure

The sample is received and weighed. 2N HCl is added to remove the impurities from the surface of the sample. The sample is immersed in deionized water to inhibit the HCl action. Acetone is used to dry the sample, and the clean, dry sample is weighed.

Sample Dissolution

The sample is placed in a graphite-bottom beaker and dissolved in 100 mL of 16N HNO_3 – 0.5M HF. The solution is boiled until 20 mL of solution remains. To the beaker, 50 mL of 1N HNO_3 is added, and the solution is boiled again. These two steps serve to ensure the complete dissolution of the sample, to reduce the volume of the solution, and to place the uranium in the 6+ valence state. The solution is transferred quantitatively to a tared poly volumetric flask for weighing. A complete transfer of the sample is ensured using 30 mL of HNO_3 as a rinse solution.

Sample and Spike Combination and Chemistry

An empty 16-mL culture tube, cap and holder are weighed. About 1 mL of spike solution is added to the culture tube, and it is reweighed. The gross minus tare serves to give the net weight of the spike. Add about 1 mL of the sample solution to the culture tube and reweigh. Subtracting the previous weight from this weight yields the weight of the sample solution added. The culture tube is mixed vigorously to obtain an isotopically homogeneous mixture. $Al(NO_3)$–$9H_2O$ is added as a salting agent to increase the solubility of the uranyl nitrate complex in the hexose. To ensure that all of the uranium is in the 6+ valence state, six drops of $KMnO_4$ are added to the culture tube. Following the addition of 2 mL of hexose the uranium is extracted from the aqueous phase to the organic phase. About 1.5 mL of the organic phase is transferred to a new culture tube, and 1 mL of H_2O_2 is added to precipitate out the uranium. Five mL of a 1:1 acetone–water mix is added to clean and dry the precipitate. The precipitate is applied directly to the filament.

Mass Spectrometric Evaluation

The solution is placed onto the sample filament of a triple filament assembly, and the filament is sent through a preprogrammed baking process to dry the filament and cre-

ate an oxide. The source carriage is installed in the mass spectrometer, and the ionization current is set to 4.0 amps for 15 minutes for remote removal of the nitrates. The sample current is then set to 1.5 amps for 15 minutes to remove the nitrates and phosphorus. The sample current is increased to 1.75 amps, and the ionization current is increased to 5.0 amps to determine the atom ratios.

Equations Used for Isotopic Fraction Determination

The basic concept used to determine the isotopic and elemental fractions of a sample is the conservation of number of atoms of a particular isotope (we assume that there are no significant nuclear reactions or decay occurring). When a spike solution of known concentration is mixed with a sample solution, the total number of atoms of a specific isotope does not change. In the case of ANL-W uranium evaluations, the isotope used in the balance is U-233. It is assumed that no U-233 exists in the sample. Therefore, it is possible to write the following balance for this isotope:

Number of U-233 atoms in spike = Number of U-233 atoms in mixture

The number of atoms in the spike can be determined because both the weight and the composition of the spike are known. The number of atoms in the mixture is, therefore, also known. The following relationships are then used to determine the weight fraction of uranium in the original sample:

$$\left(C_{sol_u}^{sam} M_{mix}^{sam} + C_{sol_u}^{Spike} M_{mix}^{Spike}\right) W_{U233}^{mix} = C_{sol_u}^{Spike} M_{mix}^{Spike} W_{U233}^{Spike}$$

(Equation 9)

and

$$C_U^{sam} = C_{sol_u}^{sam}\left[\left(M_{sol}^{sam} - M_{sol_t}^{sam}\right)/\left(M^{sam} - M_t^{sam}\right)\right]$$

(Equation 10)

where

$C_{sol_u}^{Spike}$ is the concentration of uranium in the spike,

$C_{sol_u}^{sam}$ is the concentration of uranium in the sample,

M_{mix}^{sam} is the mass of sample solution in the mixture,

M_{mix}^{Spike} is the mass of the spike solution in the mixture,

$M_{sol}^{sam} - M_{sol_t}^{sam}$ is post-dilution sample mass,

$M^{sam} - M_t^{sam}$ is the predilution sample mass,

W_{U233}^{mix} is the weight fraction of U-233 in the mixture,

W_{U233}^{Spike} is the weight fraction of U-233 in the spike, and

C_U^{sam} is the concentration of uranium in the sample (grams of uranium/gram of sample).

The weight fraction of U-233 in the mixture can be determined from the uncorrected atom ratios (those read directly from the instrument) and the mass spectrometer discrimination bias using the following equation:

$$W_{U233}^{mix} = \frac{R35\left(\frac{1}{1+2B}\right)233}{R35\left(\frac{1}{1+2B}\right)233 + R45\left(\frac{1}{1+B}\right)234 + 235 + R65(1+B)236 + R85(1+3B)2}$$

where

R35 is the uncorrected atom ratio of U-233 to U-235 in the mixture,

R45 is the uncorrected atom ratio of U-234 to U-235 in the mixture,

R65 is the uncorrected atom ratio of U-236 to U-235 in the mixture,

R85 is the uncorrected atom ratio of U-238 to U-235 in the mixture, and,

B is the mass spectrometer mass discrimination bias.

Therefore, all of the quantities in Equation 10 are known except for the concentration of uranium in the sample solution, which can therefore be determined. The following form of the equation can be used to do this:

$$C_U^{sam} = \frac{\left(M_{sol}^{sam} - M_{sol_t}^{sam}\right) C_{sol_u}^{Spike}\left(M_f^{tube} - M_t^{tube}\right)\left(W_{U233}^{Spike} - W_{U233}^{mix}\right)}{\left(M^{sam} - M_t^{sam}\right)\left(M_{f'}^{tube} - M_f^{tube}\right) W_{U233}^{mix}} \text{ tranfr}$$

(Equation 11)

where

tranfr is the fraction of sample solution transferred from the graphite beaker to the flask,

$M_f^{tube} - M_t^{tube}$ is the mass of the spike solution in the mixture, and

$M_{f'}^{tube} - M_f^{tube}$ is the mass of the sample solution in the mixture.

From this, the concentration of uranium in the sample can be directly determined. It must be emphasized that this equation is only correct if there is no U-233 in the sample. The isotopic concentration of sample is determined from the equations:

$$C_{U23X}^{sam} = C_U^{sam} \frac{\left[RX5\left(1+(X-5)B\right) - \frac{233}{23X} W_{23X}^{Spike}\left(\frac{R35}{1+2B}\right)\right]23X}{\frac{234R45}{1+B} + 235 + 236R65(1+B) + 238R85(1+3B) - \left(\frac{233R35}{1+2B}\right)\sum_{y=4,5,6,8,} W_{U23Y}^{Spike}}$$

(Equation 12)

$$C_{U23X}^{sam} = C_U^{sam} \frac{\left[RX5\left(\frac{1}{1+(5-X)B}\right) - \frac{233}{23X} W_{23X}^{Spike}\left(\frac{R35}{1+2B}\right)\right]23X}{\frac{234R45}{1+B} + 235 + 236R65(1+B) + 238R85(1+3B) - \left(\frac{233R35}{1+2B}\right)\sum_{y=4,5,6,8,} W_{U23Y}^{Spike}}$$

(Equation 13)

where

C_{U23X}^{sam} is the concentration of isotopic U-23X in the sample,

RX5 is the ratio of isotope U-23X to U-235,

B is the mass spectrometer discrimination bias, and

W_{U23X}^{Spike} is the weight fraction of isotope U-23X in the spike.

Equation 12 is used if X is greater than or equal to 6. Equation 13 is used if it is less than 5.

Linear Propagation of Errors Analysis

The propagation of errors is performed using Equation 1, given in the introduction. A list of all basic parameters, including their standard deviations and the method with which the standard deviations are determined, is given in Table 1 (facing page). The confidence intervals for the standard deviation are, in some instances, quite large or unknown. When the numbers are placed in Equation 1, a total standard deviation is obtained for each of the responses (i.e., the elemental fraction of uranium and the uranium isotopic fractions).

To evaluate the derivatives in the error propagation equation, nominal values of all parameters are used. These are obtained from typical samples. If the actual values vary significantly from the nominal values, it is necessary to recalculate the standard deviations. A numerical differentiation scheme is used in the evaluation of the derivatives. The change in the parameter is made small compared to the standard deviation to ensure that a true derivative is being calculated. This change is then made large to ensure that no significant nonlinearities are present and that the use of the linear propagation of errors is valid.

Correlations were considered in the analysis. It is possible to bound the contribution from correlation. In most cases, this bound is negligible. Where it is not negligible, it is possible to treat the impact in a conservative manner (either increasing or decreasing the standard deviation).

Results

The results of the study are summarized in Table 2. Here, the long- and short-term systematic and the random error standard deviations are given. These values can be used with a code such as MAWST [ref. 3] to determine the overall material balance standard deviation.

The confidence intervals for the standard deviations quoted above are quite large for several reasons. First, the standard deviations of the input parameters are not well-known. This uncertainty may result in a large uncertainty in the final answer. Second, correlations among the parameters are not well known. As a result, the second term on the right hand side of Equation 1 is not well characterized. Third, as for any error propagation analysis, there inevitably will be sources of error which are not described in this article. The magnitude of the error introduced by these unknown sources is, of course, unknown.

Conclusions and Further Work

This article presents a method for determining the error involved in a mass spectrometric evaluation. The method used involves propagating the basic errors and assuming a linear model. The standard deviations obtained may be in error due to the large uncertainty in the standard deviation of the basic parameters.

It is possible to compare the standard deviations obtained above with those obtained through repetitive measurements. Unfortunately, when repetitive measurements are performed, there is no way to guarantee that the sample is homogeneous, and therefore a sampling error is also introduced. Also, repetitive measurements do not describe the total error; they do not detect biases. A small number of these measurements have been performed at ANL-W, and they yield errors estimates that are consistent with the numbers given above.

The values of standard deviations presented here have associated errors. Further investigation of the basic errors to pin down their standard deviations is necessary. Knowledge

Table 1: Basic parameters used in the analysis

Variable	Definition*	Standard Deviation	Determination Method
M_{met}^{NBS}	Gross mass of metal and container	3.00e-4 g	Mettler
$M_{met_t}^{NBS}$	Tare mass of metal container	3.00e-4 g	Mettler
M_{dil}^{NBS}	Gross mass of dissolved sol'n, container	3.00e-4 g	Mettler
$M_{dil_t}^{NBS}$	Tare mass of solution container	3.00e-4 g	Mettler
Pur^{NBS}	Weight fraction of U in NBS standard	1.7e-4	NBS
R85NBS	Atom ratio U-238/U-235 certified by NBS	0.1%	NBS
R85ANL	Atom ratio U-238/U-235 by ANL-W	5.0e-4	Data
R48	Atom ratio U-234/U-238 for NBS960	1.7e-6	Data
R58	Atom ratio U-235/U-238 for NBS960	6.1e-6	Data
R43	Atom ratio U-234/U-233 for U-233 spike	1.6e-6	Data
R53	Atom ratio U-235/U-233 for U-233 spike	3.1e-6	Data
R63	Atom ratio U-236/U-233 for U-233 spike	2.2e-6	Data
R83	Atom ratio U-238/U-233 for U-233 spike	6.5e-6	Data
R38'	Atom ratio U-233/U-238 for 1:1 mixture	2.0e-3%	Data
R48'	Atom ratio U-234/U-238 for 1:1 mixture	1.5e-2%	Data
R58'	Atom ratio U-235/U-238 for 1:1 mixture	2.5e-3%	Data
R68'	Atom ratio U-236/U-238 for 1:1 mixture	7.0e-2%	Data
R35	Atom ratio U-233/U-235 in sample	2.00e-3	Dara
R45	Atom ratio U-234/U-235 in sample	5.00e-3	Data
R65	Atom ratio U-236/U-235 in sample	1.00e-2	Data
R85	Atom ratio U-238/U-235 in sample	2.00e-3	Data
M_{sol}^{sam}	Gross weight of sample solution	3.00e-4 g	Mettler
$M_{sol_t}^{sam}$	Tare weight of sample solution container	3.00e-4 g	Mettler
M^{sam}	Gross weight of sample, container	3.00e-4 g	Mettler
M_t^{sam}	Tare weight of sample container	3.00e-4 g	Mettler
M_t^{tube}	Tare weight of culture tube	3.00e-4 g	Mettler
M_f^{tube}	Culture tube and spike solution weight	3.00e-4 g	Mettler
$M_{f'}^{tube}$	Culture tube, spike and sample weight	3.00e-4 g	Mettler
tranfr	Transfer fraction from graphite beaker	1.00e-3	Expert

*Atom ratios are instrument-determined ratios before correction with the mass discrimination bias.

of the correlation structure is also necessary to tighten the confidence bounds for the standard deviations.

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Table 2: Standard deviations for the various responses

Response	Long-term Systematic Standard Deviation	Short-term Systematic Standard Deviation	Random Standard Deviation
Sample U Fraction	0.080%	0.055%	0.277%
U-234 content	0.066%	0.047%	0.530%
U-235 content	0.032%	0.016%	0.136%
U-236 content	0.001%	0.011%	1.011%
U-238 content	0.068%	0.034%	0.283%

Analysis of Active Well Coincidence Counter Measurements

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Abstract

Using a comprehensive but simple physical model, an algorithm is developed, depending only on the totals *(single count rate) and* reals *(coincident count rate). This algorithm is applied to active well coincidence counter (AWCC) measurements of high enriched uranium (HEU) material and provides a good agreement between declared and measured values of the HEU content of samples.*

1. Introduction

In several recent papers, a physical/mathematical model was developed and elaborated which described the multiplicative processes in samples containing fissile material from a general statistical viewpoint, starting with the basic underlying physical phenomena.[1,2] The results of this model agreed with the established picture used in standard high-level neutron coincidence counter (HLNCC) measurements,[3,4] but considerably extended them and allowed a more detailed interpretation of the underlying physical mechanisms and of the higher moments of the neutron counts.

On the other hand, two experimental papers used the first, second and third factorial moments, measured by a multiplicity counter with the AWCC to estimate the U^{235} content of the sample.[5,6] The analyses presented suggested that the original superfission model may not be adequate to describe the AWCC system and developed empirical correction factors and possible varying coupling factors to represent the data.

The present paper examines some recent measurements made at Y-12 (Oak Ridge) using the AWCC, in the light of the general formalism indicated initially. The results show internal consistency under a variety of conditions and provide a good agreement between experiment and theory.

2. Experimental Data

In the AWCC, an active neutron source (AmLi) is introduced into the end plugs of a detector well to induce fission reactions in U^{235} samples. The coincidence neutrons from these induced fission reactions are then a measure of the U^{235} mass in the sample.

The data examined and analyzed below were as follows:

Samples containing crushed, highly enriched (greater than 90%) U^{235} metal pieces, in various irregular configurations, were inserted into the AWCC (with an AmLi source), and the totals, T (total single count rate), and reals, R (real coincidence count rate), were measured. This process was repeated a number of times with changed amounts of the sample masses. Corresponding to each measurement,* T_0 and R_0, the background count rates were measured (without a sample present). R is determined as the difference of the measured coincidences, R + A, and the accidental coincidences, A.

3. Theoretical Background

The following discussion does not consider other instrumental effects relating to the measurements considered. Thus, questions of dead-time and die-away-time are not dealt with here; these factors are important and have been discussed elsewhere, but they are not germane to the present approach and are presumed to have been corrected for. This paper is largely concerned with the physical processes which underlie the measurements, and their explication.

Consider the sample (in the system) to be irradiated by an effective source S, which is related to the external AmLi (or other) source S_{ext} by the relation

$$S = fS_{ext},$$

where f is a factor depending on the configuration and constitution of the experimental apparatus.

The totals, T_0, i.e., the singles count-rate of the source

* Actually, this was done for each group for three or four measurements.

alone, is then

$$T_0 = \varepsilon_0 S \tag{1}$$

where ε_0 is a proportionality constant, including the measurement efficiency.

When a sample is introduced into the well, assume that each neutron may induce a fission with probability p_f, may be absorbed (captured) in the sample with probability p_c, and, if it gets into the detector (with probability $1 - p_f - p_c$), has a probability ε of being detected. Thus the overall detection probability of each neutron is $\varepsilon(1 - p_f - p_c)$.†

Here, ε is again a proportionality constant involving the instrumental efficiency and depends on the sample configuration and constitution. Since these factors modify the neutron behavior, ε is not necessarily equal to ε_0.

Using the physical model described in Reference 1 (Equations 21 and 22 on p. 474), the totals and reals become

$$T = \frac{\varepsilon(1 - p_f - p_c)}{1 - p_f \upsilon_1} S \tag{2}$$

$$R = \frac{\varepsilon^2(1 - p_f - p_c)^2}{2(1 - p_f \upsilon_1)^2} \frac{p_f}{(1 - p_f \upsilon_1)} \upsilon_2 S \tag{3}$$

where $\upsilon_1 = <\upsilon>$ is the average number of neutrons emitted per induced fission, $\upsilon_2 = <\upsilon(\upsilon - 1)>$ is the second factorial moment of the induced neutron distribution, etc., and S is the known source rate.

R is very sensitive to the value of p_f. In the first approximation, the probability of inducing a fission is proportional to the U^{235} mass in the sample, and this approximation remains valid for samples of similar physical and chemical characteristics and for situations in which the U^{235} mass is not too large. The approximation will not be valid, however, when the sample contains enough moderator to slow down the neutrons appreciably, nor when the amount of U^{235} becomes so large *and* when surface to volume ratio of the sample becomes small so that self-shielding effects become important

Assuming for convenience and perspicuity that $p_c = 0$,

$$T = \varepsilon M S \tag{4}$$

$$R = \frac{\varepsilon^2 M^2 (M - 1) S <\upsilon(\upsilon - 1)>}{2(\upsilon_1 - 1)}, \tag{5}$$

† A more elaborate treatment would introduce separate sets of parameters for the source and the induced neutrons, see Lu and Teichmann, 1992, ref. 1.

where M is given by

$$M = \frac{1 - p_f}{1 - p_f \upsilon_1}. \tag{6}$$

Introducing

$$\delta = \frac{\varepsilon_0 - \varepsilon}{\varepsilon},$$

one has

$$M = \frac{T}{T_0}(1 + \delta), \tag{7}$$

which indicates, that to a first approximation, T/T_0 can be used as an estimate of the leakage multiplication factor M.

Thus,

$$\frac{R}{\left(\frac{T}{T_0}\right)^2} = \frac{1}{2}\varepsilon_0^2 S \upsilon_2 \left(\frac{T(1 + \delta)}{T_0} - 1\right)(1 + \delta)^2, \tag{8}$$

or, in a more convenient form,

$$\frac{R}{\left(\frac{T}{T_0}\right)^2} = \left(\frac{\varepsilon_0^2 \upsilon_2 S}{2(\upsilon_1 - 1)}\right)(M - 1). \tag{9}$$

Noting the remarks preceding Equation 4, this suggests a close correlation between M – 1 and m, which is pursued below.

4. Multiplication-Fissile Mass Correlation

Equation 9 leads one to consider the question of developing a correlation between the multiplication factor, M, and the fissile (U^{235}) mass m. Since M = 1 for m = 0, it is convenient to search for the correlation in the form

$$M - 1 = f(m). \tag{10}$$

Referring to Equation 9, one finds that here

$$f(m) = C \frac{R}{\left(\frac{T}{T_0}\right)^2}. \tag{11}$$

Figures 1, 2, and 3 show plots of the measured mass,

$$C\frac{R}{\left(\frac{T}{T_0}\right)^2}$$

versus, m, the declared mass of U^{235} for three sets of experiments.** (The corresponding consolidated data are shown in Figure 5, *Consolidated AWCC Data.*) The determination of the appropriate calibration constant(s) C is shown in Figure 4, in which

$$\frac{R}{\left(\frac{T}{T_0}\right)^2}$$

is plotted against m for the three cases (cases 2 and 3 were combined because of their similarity)

Thus for these sets of experiments there is an excellent *linear* correlation between the leakage multiplication factor M and the declared fissile (U^{235}) mass m, and thus for the measured U^{235} mass and the declared U^{235} mass. For similar experimental configurations and samples, a similar linear relationship may be expected, though the constant C may vary, depending, as it does, on the source strength and the parameters relevant to the induced fission probability. In the present case, the difference in the values of C can be attributed to a change in the geometric configuration of the experiment (In the first set of measurements the effective source strength was increased due to the changed geometry).

The final figure, Figure 6, based on the first set of experiments, shows relations between the calculated and the declared mass based on different measurement models.

It should be noted that the linearity observed here, while heuristically plausible, is by no means the consequence of general physical requirements. In the case of the HLNC, a more complicated relationship applies, though the calculated mass is proportional to the declared mass when the latter is small. It will be of interest to determine the correlation for differently constituted samples and for larger U^{235} masses.

** The combined data for the three sets is shown in the table "Consolidated AWCC Data." The column labeled "calculation" is the quantity $R/(T/T_0)^2$.

Acknowledgment

We are greatly indebted to William Higinbotham for a number of penetrating discussions about these experiments and their interpretation, and for a careful and critical review of the analyses.

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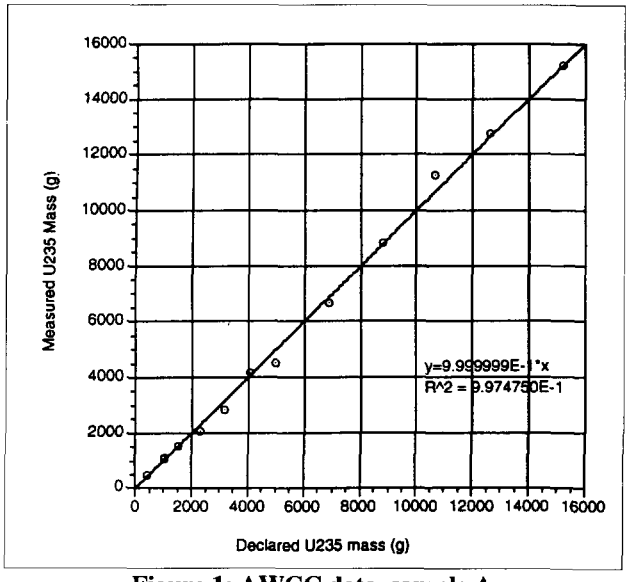

Figure 1: AWCC data, sample A

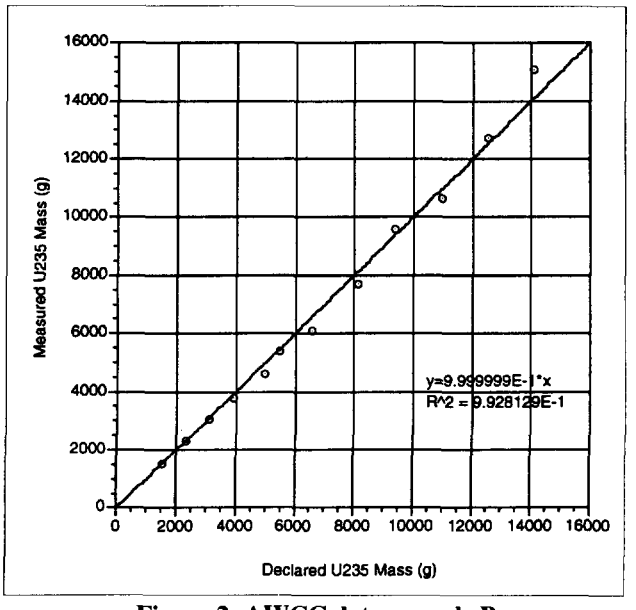

Figure 2: AWCC data, sample B

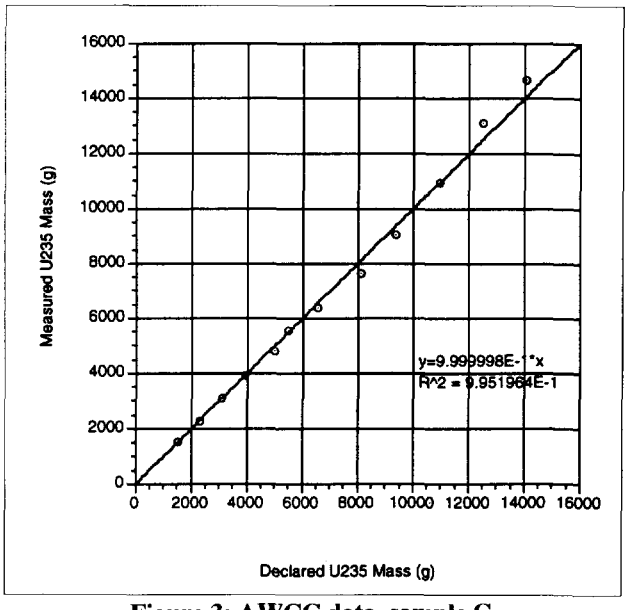

Figure 3: AWCC data, sample C

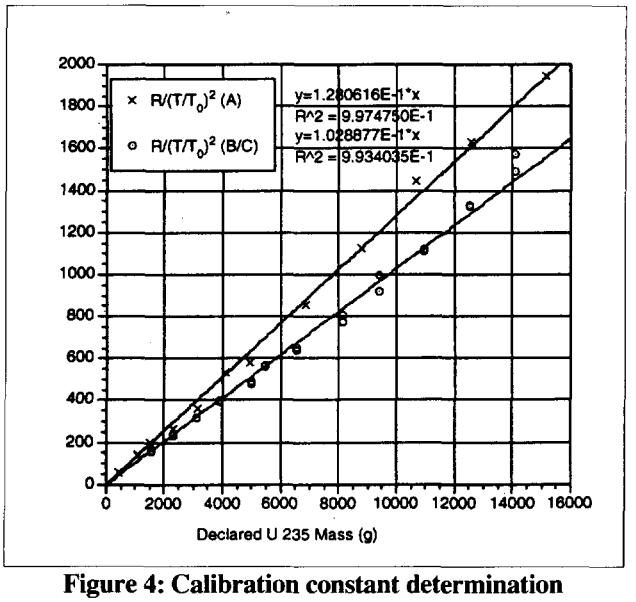

Figure 4: Calibration constant determination

Consolidated AWCC Data

ID	Date	T0	T-T0	T/T0	R	Declared U Mass	Calculation	Declared U-235 Mass	Measured U-235 Mass
1452-1	930422	10894	3263	1.2995	3278	16309	1941	15193	15157
1452-3	930422	10894	2794	1.2565	2571	13557	1629	12628	12717
1452-4	930422	10894	2525	1.2318	2184	11465	1439	10680	11240
1452-5	930423	10896	2085	1.1914	1592	9433	1122	8787	8759
1452-6	930423	10954	1653	1.1509	1125	7385	849	6879	6632
1452-7	930423	10954	1159	1.1058	700	5340	572	4974	4470
1452-8	930423	10954	1147	1.1047	645	4400	529	4099	4127
1452-9	930423	10973	843	1.0768	415	3415	358	3181	2795
1452-10	930423	10973	678	1.0618	290	2498	257	2327	2009
1452-11	930426	10922	497	1.0455	210	1645	192	1532	1500
1452-12	930426	10922	339	1.031	137	1168	129	1088	1006
1452-12	930426	10922	337	1.0309	142	1168	134	1088	1043
1452-13	930426	10922	177	1.0162	56	464	54	432	423
1452-13	930426	10922	186	1.017	55	464	53	432	415
C1-1	930806	11007	455	1.0413	168	1681	155	1566	1488
C1-2	930806	11007	631	1.0573	263	2525	235	2352	2262
C1-3	930806	11007	762	1 .0692	356	3362	311	3132	2990
C1-4	930806	11007	976	1.0887	461	4212	389	3923	3739
C1-5	930806	11007	1138	1.1034	576	5378	473	5010	4547
C1-6	930806	10997	1334	1.1213	700	5891	557	5487	5349
C1-7	930806	10997	1453	1.1321	805	7060	628	6576	6034
C1-8	930806	10997	1732	1.1575	1065	8749	795	8150	7636
C1-9	930806	10997	2015	1.1832	1388	10097	992	9405	9530
C1-10	930806	11014	2235	1.2029	1598	11778	1104	10971	10611
C1-11	930806	11014	2503	1.2273	1989	13458	1320	12536	12686
C1-12	930806	11014	2779	1.2523	2456	15138	1566	14101	15045
C2-1	930806	11007	401	1.0364	161	1681	150	1566	1474
C2-2	930806	11007	604	1.0549	253	2524	227	2351	2233
C2-3	930806	11007	803	1.0729	359	3701	311	3447	3063
C2-4	930806	11007	980	1.0891	468	4202	395	3914	3883
C2-5	930806	11007	1160	1.1054	593	5050	485	4704	4769
C2-6	930806	10997	1314	1.1194	699	5887	558	5484	5488
C2-7	930806	10997	1491	1.1356	830	7068	643	6584	6327
C2-8	930806	10997	1692	1.1539	1024	8742	769	8143	7559
C2-9	930806	10997	1919	1.1745	1260	10425	913	9711	8980
C2-10	930806	11014	2274	1.2065	1616	11776	1110	10969	10917
C2-11	930806	11014	2462	1.2235	1985	13459	1326	12537	13040
C2-12	930806	11014	2757	1.2503	2319	15140	1483	14103	14584

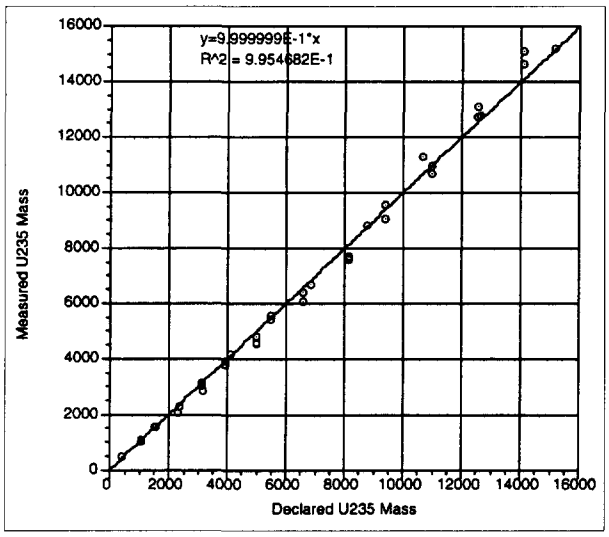

Figure 5: Consolidated AWCC data

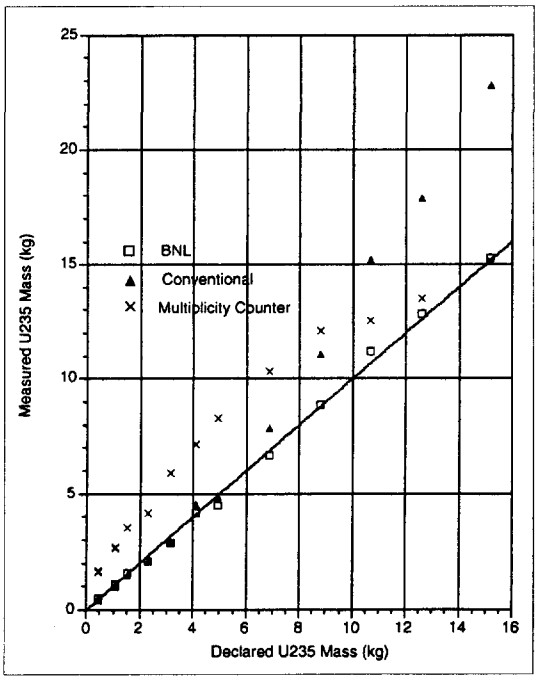

Figure 6: Comparison of different models

International Status and Trends for Spent Fuel Management

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Abstract

Spent fuel management encompasses all the activities associated with handling, transport, storage, processing and disposal of spent nuclear fuel following its discharge from the reactor.

The first part of this paper describes the various approaches to the back-end of the nuclear fuel cycle. The second part reports data on the amount of spent fuel discharges from nuclear power reactors and compares these with the available reprocessing capacities. The difference between the two values is the minimum amount of fuel to be stored.

Various types of interim storage facilities are described together with a summary table for the away from reactor fuel storage capacities. A separate table for the spent fuel inventory in the former CMEA countries and in the Republics of the former Soviet Union is provided.

Activities of the International Atomic Energy Agency related to management of spent fuel from power plants and reactors, including the develiopment of international safety guidelines and the initiation of the Irradiated Fuel Management Advisory Program, are described.

Introduction

Operation of nuclear power plants or research reactors generates spent nuclear fuel for which suitable management arrangements must be made. The amount of spent fuel to be stored continues to increase. Management of this spent fuel has always been one of the most important tasks in the nuclear fuel cycle, and it is still one of the most vital problems common to all countries with nuclear reactors. Spent fuel management encompasses all the activities associated with handling, transporting, storing, processing and disposal of spent nuclear fuel following its final discharge from the reactor. The time-scale of commitment to the safe management of spent fuel is many decades.

Spent Fuel Management Options

Once the spent fuel has been discharged from the reactor, it is normally cooled for some period of time in a water pool or equivalent storage system. This allows the radioactive decay of the shorter-lived radionuclides and an associated reduction in the emission of heat resulting from the decay processes. Because most at-reactor storage pools have limited capacity, usually only for a few years of arisings, it soon becomes necessary for a country to decide on a national policy for safe management of the spent fuel. There are three basic variations on the path to final disposal of the spent fuel arisings:

- the closed fuel cycle (reprocessing),
- the once-through fuel cycle (direct disposal), and
- deferral (interim storage).

The Closed Fuel Cycle

In this process, after initial cooling the spent fuel is transported to a reprocessing facility where the residual fuel values (uranium and plutonium) are separated from fission products and other actinides. The recovered uranium and plutonium are returned to the fuel cycle for reuse in subsequent fuel fabrication as uranium or mixed oxide (MOX) fuel. The fission products and remaining actinides as well as any medium and low level wastes from reprocessing must be solidified and encapsulated or otherwise processed for disposal.

The Once-Through Fuel Cycle

In this case the spent fuel is transported to the final disposal facility where it is suitably conditioned and packaged for the disposal site environment. It is placed in the repository without any recovery of residual fuel values.

However, since the development of a repository — either for the spent fuel or for the high-level wastes — is a lengthy undertaking, which involves selection of suitable geology and location and licensing, it is necessary to store the spent fuel safely for some extended period until such time as repositories are available for service. No country currently has a licensed site, and projections suggest this will be after 2010.

Deferral/Interim Storage

Delays in the implementation of the fuel reprocessing option in some countries, the complete abandonment of this option in other countries and delays in the availability of final spent fuel disposal in almost all countries has led to increasingly long periods of interim spent fuel storage. This "wait and see" approach gives more time and freedom to evaluate the available options and to select the most suitable technology. The problem of spent fuel management has therefore increased in importance for many countries.

The requirements for interim storage of spent fuel are derived from a number of considerations associated with the future management options of reprocessing or disposal.

In the closed fuel cycle option, further storage capacity may be required to match the arisings of fuel with the available reprocessing plant capacity. With respect to the once through cycle, storage is required for materials until the final repository has been engineered and is in service. Clearly for deferral option, the availability of adequate interim storage is a key element.

Because of its importance it is necessary to have a clear understanding of storage. Storage is not disposal, because storage retains the ability to retrieve. It is a fundamental principle of storage that, whatever technical proposal is adopted, it does not prejudice the ability to meet the overall policy requirement. For example, where interim storage is to be followed by direct disposal, the physical condition of the fuel at the end of the storage phase must meet the acceptance criteria of the repository.

Table 1 shows the spent fuel management approaches selected by different countries for their nuclear power plants.

Table 1: Spent fuel management approaches* selected in different countries

	Deferred decision	Direct disposal	Reprocessing
Argentina	x	-	x
Belgium	-	-	x
Brazil	-	-	x
Bulgaria	x	-	x
Canada	-	x	-
China	-	-	x
Czech Republic	x	-	x
Finland	x	x	x
France	-	-	x
Germany	-	x	x
Hungary	x	-	x
India	x	-	x
Italy	x	-	x
Japan	-	-	x
Korea, Rep. of	x	-	-
Lithuania	x	-	-
Mexico	x	-	-
Netherlands	-	-	x
Pakistan	x	-	-
Russia	-	x	x
Slovak Republic	x	-	x
Slovenia	x	-	-
South Africa	x	-	-
Spain	x	x	x
Sweden	-	x	
Switzerland	x	-	x
UK	x	-	x
Ukraine	x	x	x
USA	-	x	-

* Some countries have different spent fuel management approaches for different fuel types. In some countries, one spent fuel management approach is presently being followed but future options applying different approaches are being evaluated.

Spent Fuel Arisings, Amounts of Spent Fuel Being Stored

Power Plants

The current and projected quantities of fuel being discharged from power plants world-wide are illustrated in Figure 1 (next page) for the major generators. While most of this material will be stored for an extended period prior to final disposal, a significant amount is already committed to being reprocessed, thereby reducing the cumulative quantity of spent fuel requiring storage and disposal. The projected amount of spent fuel that will require storage and eventual disposal is illustrated in the second column of Figure 1 after subtraction of that amount of spent fuel that could be accommodated by the available reprocessing capacity. Of course, if material recovered from reprocessing reenters the fuel cycle, it will reappear again as spent fuel requiring repro-

cessing or disposal and will affect the amount of new uranium entering the cycle.

Reprocessing of spent fuel involves the separation of fissile and fertile material, mainly Pu and U, from fission products and the recovery of the fissile isotopes for recycling in FBRs or thermal reactors. Reprocessing is now a proven technology. Commercial reprocessing services are now available to reactor operators. The projected reprocessing capacities according to fuel type are summarized in Table 2. The total capacity for all fuel types in 1992 amounted approximately to 4,015 t HM. All reprocessing plants use advanced Purex technology. No major problems are anticipated in dealing with fuels of high burnup or with MOX fuel. UP-800 in France, THORP in the United Kingdom and the Rokkasho Mura plant in Japan are by design capable of reprocessing high burnup fuel up to 45,000 MWd/t.

The UP2 reprocessing plant in France is being modified to double its capacity to 800 t HM and is now scheduled to start up at the doubled capacity in 1994–1995. Construction of the Thermal Oxide Reprocessing Plant (THORP) at Sellafield in the United Kingdom was complete in 1992, but commissioning was delayed.

The total amounts of commercial spent fuel which have been reprocessed up to through 1992 is about 46,000 t HM (34,300 t HM GCR, 11,700 t HM LWR and 40 t FBR fuel).

In 1992, the spent fuel arising from all types of reactors in nuclear power plants amounted to about 10,000 t HM, giving an estimated cumulative total of more than 135,000 t HM. Of this, about 90,000 t HM of spent fuel is stored at present. The quantity of accumulated spent fuel is more than 20 times the current total annual reprocessing capacity. According to the projections, the annual spent fuel arising will grow gradually from about 10,000 t HM in 1992 to over 11,000 t HM in 2000. The projected cumulative amount of spent fuel generated by the year 2000 will reach 225,000 t HM.

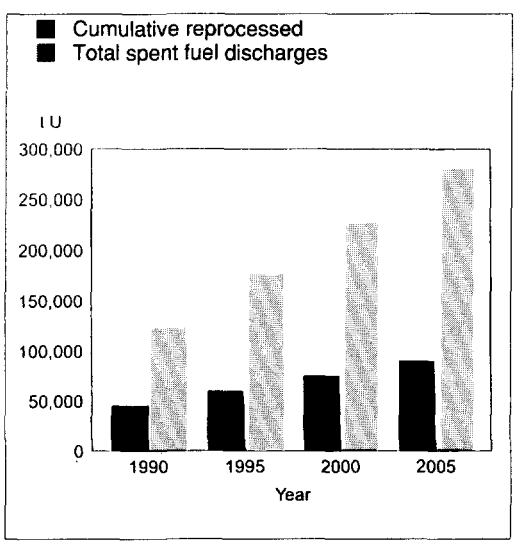

Figure 1: Amounts of spent fuel discharged and to be stored

Assuming that part of it is reprocessed, the amount to be stored by the year 2000 is projected to be about 150,000 t HM.

Since the first large-scale final repositories for disposal of spent fuel are not expected to be in operation before the year 2010, interim storage will be the primary option for the next 20 years.

Research Reactors

A concern to many of our member states is the question of spent fuel from the research reactors. At present there are 325 research reactors operating all over the world. Out of these about 80 units are operating, 10 are under construction and 9 already shut down in 34 developing countries. Figure 2 shows the age distribution of these reactors. As can be seen from it, most of the reactors were constructed 25 to 35 years ago. At the time of construction it was assumed in most instances that the irradiated fuel would be shipped back to the country of origin. Since 1988 this has not always proved to be possible, and in many countries highly enriched, high burn-up fuel is being stored in facilities not originally designed for long-term fuel storage and lacking the auxiliary systems required for safe operation and monitoring.

For research reactors, the IAEA does not have a comprehensive database on the amount of spent fuel in storage at present. To rectify this situation, a questionnaire has recently been sent to the operators of the research and test reactors and the responses received are being evaluated.

The United States has exported more than 25,000 kilograms of highly enriched uranium (HEU). Of this, about 17,500 kg is currently in use or stored as spent fuel in 51 countries. Most of the exported HEU went to the 12

	Fuel type [a]	1995	2000	2005	2010
France	GCR	600	0	0	0
	LWR	1,600	1,600	1,600	1,600
	FBR	5	5	5	5
India	PHWR, RR	200	600	600	600
Japan	LWR	100	900	900	900
Russia	LWR	400[b]	400[b]	400[b]	400[b]
UK	GCR	1,500	1,500	1,500	1,500
	LWR	1,200	1,200	1,200	1,200
	FBR	10	10	10	10
Total		**5,615**	**6,215**	**6,215**	**6,215**

[a] GCR: gas cooled reactor;
LWR: light water reactor;
FBR: fast breeder reactor;
PHWR: pressurized heavy water reactor;
RR: research reactor.

[b] The completion of a reprocessing plant at Krasnoyarsk of 1,500 t HM/a capacity has been postponed.

Table 2: Projected reprocessing capacities in the world (t HM/a)

EURATOM countries (85%), the remaining amount to 39 non-EURATOM countries. About 25 research reactors in 11 countries have been exported by the Soviet Union in the past. Through the end of 1993, there was no return of spent fuel from these either.

As a result many operators of research reactors find themselves in a crisis situation because of the spent fuel management problems. This is particularly the case in several Western European countries where operating license extensions are tied to a successful resolution of spent fuel problems. The crisis has been precipitated by the cessation of take-back of research reactor fuels by the countries where they were originally enriched (mainly the United States and Russia). The crisis has been exacerbated by the Reduced Enrichment for Research and Test Reactors (RERTR) program which has left many pools at research reactors filled with HEU and left to cope with a greater throughput of fuels of lower enrichment. Although there are encouraging signs that both the United States and Russia will renew their take-back of research reactor fuels, any protracted delay in the implementation of these policies could lead to the closure of important research facilities.

Spent Fuel Storage Facilities for Nuclear Power Plants

The requirements for interim storage of spent fuel depend on a number of considerations associated with the management option selected. In the closed fuel cycle option, further storage capacity may be required to match the arisings of spent fuel with the available reprocessing plant capacity. For the once-through cycle, storage is required until the final repository has been engineered and is in service. Clearly, for deferred decisions the availability of adequate interim storage is also a key element.

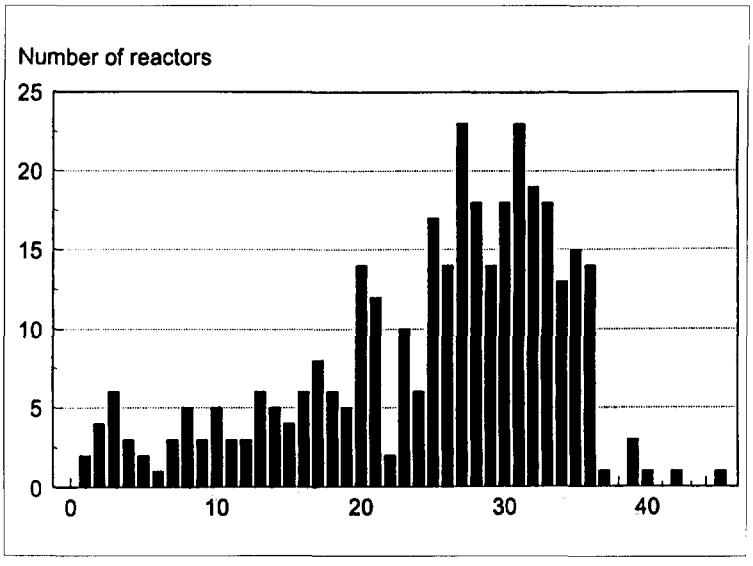

Figure 2: Age distribution of research reactors in 1993

Various types of wet and dry storage facilities are in operation, or being considered by our member states.

Wet storage is by far the most common form at reactors and at reprocessing plants. This is not surprising because most large commercial plants have light (or heavy) water reactors in which the fuel is designed to dwell in water for long periods without deterioration and which provide the necessary cooling after discharge. Historically interim storage prior to reprocessing has been wet, but there is no technical reason for this and dry storage has been used and may be used more in future.

Most strategies now consider dry storage for the longer term because of its inherent passive nature and low operating costs. Dry spent fuel may also be more amenable to conditioning for disposal. Trends in the future are therefore more likely to be for dry storage during the interim period between discharge from reactor pools and the next phases of fuel management. The exception might be for the current generation of reprocessing plants which are based on wet interim storage. Most current and future plant will not have life time capacity incorporated within the initial pool design for safety and economic reasons and further storage will be necessary during plant operating life.

To update our spent fuel database, we at the IAEA regularly send out questionnaires. A constant problem in the evaluation of the responses is how to define the type of a spent fuel storage facility. For this article and in our publications we decided to use the following 3 classes:

- At-reactor storage,
- Away-from-reactor storage, on the reactor site, and
- Awary-from-reactor storage, off site.

At-reactor (AR) storage is that which is associated with the reactor itself as part of the original design even though it might have been enlarged or extended during reactor life. It is essentially the spent fuel pool or its equivalent in dry fuel discharge routes, and provides the initial post discharge cooling and storage which may be for a few hundred days, a few years or even the plant's operating life. It exists in almost all plants.

Away-from-reactor (AFR) storage means a storage facility which is independent of the reactor, although it may be adjacent to it on the same site or at a separate site. In the United States, the term independent spent fuel storage installation (ISFI) is used. The location of the AFR is an important consideration in spent fuel management and the terms AFR (RS) and AFR (OS) will be used to distinguish between facilities at the reactor site (RS) and those at other locations, i.e., off site (OS).

When, for various reasons, the capacity of the AR pools has to be increased, the nuclear plant operators have often first implemented various in-pool measures, e.g., compacting the storage racks, rod consolidation and double-tiering, before adding extensions to existing AR storage. This was usually more economic and may

have been the only immediate solution. Some direct extensions have been possible to AR pools at a limited number of reactors. CANDU reactors on the other hand have very large AR pools which can store nearly all lifetime arisings.

Having exhausted the AR storage capacity expansion opportunities, the practice adopted by many utilities has been to have some form of interim facility constructed, either in the form of wet or dry storage. These facilities usually are constructed as AFR(RS) storage facilities. This has often been a perfectly viable option, but usually the long term spent fuel management strategy was not considered as the generation of electricity was the priority.

Water pools have certainly been built for use as AFR(RS) storage facilities with the necessary arrangements to transfer fuel across the site. The largest AFR(OS) pools, however, are at reprocessing plants at La Hague (France) and at Sellafield (United Kingdom). These can store 5,000 to 10,000 t HM. The alternative types of storage being used or planned are dry, passive systems. These may be large, massive structures as in the case of a natural convection vault, or may be smaller, incrementally added individual units, as in the examples of the metal cask, the vertical or horizontal concrete silos, and the drywells.

Table 3 shows the AFR(RS&OS) storage capacities in operation and planned in various countries. In 1992 the capacity of the AFR storage in operation was 47,373 t HM. Of this 44,833 t HM was in wet storage and 2,540 t HM in dry storage. The figures also include the pool capacities of reprocessing facilities. Most of the fuel in the AFR storage facilities is under water and considerable positive experience exists internationally with this kind of wet storage for periods of up to 40 years. This has been proven also by the IAEA's Coordinated Research Program (CRP) on the behavior of spent fuel and storage facility components during long-term storage (BEFAST), which has been ongoing since 1979.

	In operation	Under construction or planned	Shut down/ On standby
Argentina	365		
Belgium			370
Bulgaria	600		
Canada	475	12,800	
China		500	
Czech Republic		600	
Finland	1,270		
France	15,000		
Germany	2,150	700	1,500
India	523		
Japan	140	3,000	
Korea, Rep. of		3,000	
Russia	13,100	1,900	
Slovakia	600		
Spain		5,500	
Sweden	3,000	6,000	
UK	10,350	1,200	
Ukraine	1,900		
USA	900	15,000[a]	
Total	50,373	50,800	1,870

[a] A number of U.S. reactor operators have licensed additional dry storage facilities.

Taken from the IAEA Yearbook 1993.

Table 3: Away-from-reactor storage capacities (t HM)

Nearly all countries operating nuclear power plants have increased their existing AR capacity by re-racking using neutron-absorbing materials between the assemblies, through rod consolidation or simply by better distribution of fuel in the storage pools. Such modifications have resulted in about a twofold increase in storage capacity. Further capacity increases may invoke the so-called burnup credit in calculations of the criticality of irradiated fuels. In many cases, modifications were insufficient and separate AFR storage facilities had to be constructed. Although the majority of storage facilities are of the wet type (e.g., in France, the United Kingdom, Russia and Sweden), many countries with large quantities of spent fuel have chosen or are choosing AFR dry storage (e.g., Canada, Germany, Scotland and the United States, while in Russia dry storage is being developed for RBMK fuel). This type of storage has many benefits including the possibility of passive cooling, minimal or no maintenance and a non-corrosive environment.

Changes in the politics and trading relationships of the Eastern European countries are affecting their spent fuel management policies. Russia now requires payment for services in hard currency at a "world price" level. Some legal problems also exist with the transport of Russian-origin fuel and its subsequent reprocessing in Russia. Such factors may lead to changes in the spent fuel management policy of these countries. Construction of an interim storage facility can be a temporary solution, with the options of reprocessing or direct disposal kept open. For example, in 1992 Hungary decided to build an AFR storage facility at its Paks Nuclear Plant. An independent group of experts, convened by the IAEA, helped Paks evaluate the various technologies. A dry vault type facility was chosen finally by the operator. This facility is scheduled to be ready by 1995. Experts in the Czech Republic decided to construct a dry cask storage facility at the Dukovany site. Other WWER reactor operators are also investigating the options available to them for increasing spent fuel storage time. Table 4 gives an inventory of the spent fuel from power reactors for these countries.

Recognizing the mature status of AFR technologies, changes in the basic principles are not expected in the near future. Nevertheless, factors that may influence future AFR designs are as follows:

- The impact of disposal concepts (when finalized). There may be pressure to place spent fuel in containers amenable to disposal requirements at as early a stage as

practicable to minimize the number of handling operations.

- New requirements to store higher burnup fuel and/or advanced fuel types.
- The desire of utilities to claim burnup credits against criticality considerations.
- The desire of utilities to extend storage times, which will influence inspection and maintenance requirements of AFR facilities and put more emphasis on fuel degradation mechanisms.

Highlights of Future IAEA Activities[1]

The Nuclear Materials and Fuel Cycle Technology Section is responsible for spent fuel related activities of the IAEA. This work is mostly organized through different types of meetings and resulting in a variety of publications.

Since the publication of the *Guidebook on Spent Fuel Storage* in 1984, many documents have been published as technical reports (sales publications). A list of such sales publications has recently been published. The catalogue of the sales publication is available from the IAEA secretariat without chargeupon request.

A second group of publications isknown as TECDOCs; their scope also covers a wide range of topics. These publications are also available from the Secretariat upon request and without charge.

An International Symposium on Spent Fuel Storage: Safety, Engineering and Environmental Aspects is planned for October 10–14, 1994, in co-operation with OECD/NEA. A seminar on the same subject was held in 1990, with more than 100 participants from 28 countries and four international organizations. The proceedings of this symposium will be published.

Country	AR	AFR	Total
Armenia	30	-	30
Bulgaria	320	120	440
Czech Rep.	170	(140[a])	170
Hungary	300	-	300
Lithuania	800	-	800
Russia	3,900	4,950	8,850
Slovak Rep.	140	440[b]	580[b]
Ukraine	830	1430[c]	2260

[a] This fuel is in the AFR in the Slovak Republic and will be taken back.
[b] Including the 140 t HM spent fuel from the Czech Republic.
[c] RBMK fuel.

Table 4: Spent fuel inventory from power reactors in the former CMEA countries and in the Republics of the former Soviet Union (t HM)

To improve the already good performance of existing storage facilities and, in particular, to offer advice to countries now starting the construction of such objects, the IAEA has the following programs:

- preparation of a set of safety documents to develop international guidelines on the safety of spent fuel storage.
- advisory programs on the safety of spent fuel management.

Safety Series Documents on Spent Fuel Storage

At present, three documents are being prepared on the safe storage of spent fuel from power reactors. The first is a safety guide on the design of spent fuel storage facilities, the second is a safety guide on the operation of these facilities and the third is a safety practice document on the preparation of safety analysis report for spent fuel storage.

These documents are prepared by a series of consultants' meetings, advisory group meetings, and technical committee meetings that bring together the world-renowned experts in this field and are published after repeated review by the Agency's Safety Series Review Committee. According to the current schedule, the documents will be ready for publication this year. It is expected that they will be useful to member states in establishing their national standards.

Preparations to start drafting a new safety guide on the design, operation and licensing of storage of spent fuel from research and test reactors are presently under way.

Irradiated Fuel Management Advisory Program (IFMAP)

As mentioned above, for both research and commercial reactors, irradiated fuel is being stored for longer than originally envisaged and in larger quantities. While methods of increasing the existing storage capacities, or building additional stores according to modern standards, have been developed in a small number of industrialized countries, information is not always readily accessible outside the country of origin.

In view of the diversity of fuel types, particularly in research reactors, there are benefits to be derived from impartial assessments of technological concepts, operating experience, safety and regulatory aspects of irradiated fuel management before important decisions are made concerning possible long term solutions. This information is also important for countries that have operated research reactors extensively over the last 40 years. In order to fulfill these requirements, the IAEA has started a new program, The Irradiated Fuel Management Advisory Program (IFMAP). A booklet describing IFMAP and how to request the services it offers was recently sent to member states. IFMAP will provide advice in the specific area of irradiated fuel storage and on developing national programs for member states, particularly developing countries, that request its services.

In 1990 a group of experts visited China to provide ad-

vice on the storage of spent fuel from their nuclear power plant. As mentioned above, in 1992 and 1993 an IAEA team assisted a Hungarian operator in selecting and evaluating a spent fuel interim storage option. Also in 1992, preliminary discussions were held by IAEA staff members with their counterparts in the countries in question to assist in the formulation of the spent fuel storage programs in Romania, the Ukraine and Thailand.

To assist experts from developing countries to improve the operation of their storage facilities the Agency has started to organize interregional and regional training courses. These 2–3 week courses were offered in 1993 and will be offered almost every year thereafter to all interested member states in the case of interregional courses, or to those in a selected geographical area in the case of regional courses. Separate courses will be offered for power plant and for research reactor operators.

References

1. Takáts F., Grigoriev A., and Ritchie I.G. "Management of spent fuel from power and research reactors: International status and trends," *IAEA Bulletin*, Vienna, March 1993, pp. 18–22.

Ferenc Takáts has been in the IAEA department of nuclear energy and safety since 1990. He received his degree in nuclear engineering from the Moscow Institute of Power Engineering and began his career in a power station in Hungary.

April 24–26, 1994
New Mexico Conference on the Environment, Albuquerque Convention Center, Albuquerque, N.M. *Sponsor*: The Governor of the State of New Mexico and the New Mexico Environment Dept. *Contact*: John Geddie, 1190 St. Francis Dr., P.O. Box 26110, Santa Fe, NM 87502; phone (505) 827-2850.

May 2–6, 1994
ISA/94 Philadelphia, International Conference, Exhibition & Training Program, Pennsylvania convention Center, Philadelphia, PA. *Sponsor*: Instrument Society of America. *Contact*: ISA, 67 Alexander Dr., P.O. Box 12277, Research Triangle Park, NC 27709; phone (919) 549-8411.

May 9–13, 1994
1994 Incineration Conference, Adam's Mark Hotel, Houston, Texas. *Sponsor*: University of California, Irvine. *Contact*: Lori Barnow, University of California, Office of Environment, Health & Safety, Irvine, CA 92717-2725; phone (714) 856-7066.

May 15–18, 1994
Nuclear Energy Assembly (the annual conference of the Nuclear Energy Institute), Washington Court Hotel, Washington, D.C. *Sponsor*: Nuclear Energy Institute. *Contact*: NEI Conference Office, 1776 I Street, N.W., Suite 400, Washington, D.C. 20006-3708; phone (202) 466-0246

May 22–24, 1994
WFNM 21st Annual Meeting and International Conference on Nuclear Energy, Astoria Hotel, St. Petersburg, Russia. *Sponsor*: World Nuclear Fuel Market. *Contact*: Donna Cason, administrative director, WNFM, 655 Engineering Dr., Suite 200, Norcross, GA 30092.

June 19-23, 1994
1994 ANS Annual Meeting, New Orleans Hilton, New Orleans, LA. *Sponsor*: American Nuclear Society. *Contact*: General Chair Edwin Lupberger, Entergy Corp., 225 Baronne St., New Orleans, LA 70112; phone (504) 569-4301.

July 17–20, 1994
INMM 35th Annual Meeting, the Registry Resort Hotel, Naples, FL. *Contact*: Kathleen Sweeney, INMM Headquarters, 60 Revere Dr., Suite 500, Northbrook, IL 60062; phone (708) 480-9573.

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  1. Jones F.T., Chang, L.-K. "Article Title," *Journal* 47(No. 2):112-118 (1980).
  2. Jones F.T., *Title of Book*, New York: McMillan Publishing, 1976, pp.112-118.
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