



**Division of Nuclear Chemistry and Technology**  
*American Chemical Society*

NUCL WWW Home Page – <http://www.nucl-acs.org>

**NEWSLETTER**  
**50<sup>th</sup> Anniversary Special Issue**

*Newsletter Editor: Andrew Klose*  
*Email: [andrew.klose@augie.edu](mailto:andrew.klose@augie.edu)*

**Table of Contents**

- > **ACS NUCL Division 50th Anniversary: Introduction**  
David Hobart (LANL/FSU) – Page 2
- > **From the Cold War through Nuclear Deterrence: Four Decades at the Savannah River Laboratory**  
Don Orth (SRNL) – Page 8
- > **Reflections on My 70 years in Chemistry**  
Darleane Hoffman (LBNL/LLNL/UCB) – Page 10
- > **Challenges and Rewards in Nuclear Chemistry**  
Paul Karol (Carnegie Mellon University) – Page 13
- > **Probing and Understanding the Science of the Actinide Elements**  
Richard Haire (ORNL) – Page 17
- > **So What do you get for Being a Member of the Division (1986 – Present)**  
Michael Bronikowski (SRNL) – Page 23
- > **Evolving Professional Preferences**  
Corinne Dorais (U. of Notre Dame) – Page 26
- > **History of the Division of Nuclear Chemistry and Technology's Summer Schools in Nuclear Chemistry**  
Trish Baisden (LLNL) – Page 29
- > **Reflections on 50 Years in Nuclear Science**  
Major Thompson – Page 38
- > **A Career from Petrology to Nuclear Science**  
Annie Kersting (LLNL) – Page 40
- > **From Nuclear Chemistry to Nuclear Medicine: The Role of NUCL Chemists in Medical Imaging**  
Suzanne Lapi (U. of Alabama at Birmingham) and Alan Packard (Harvard Medical School) – Page 43
- > **In Memoriam: Alan Davison 1936 – 2015** – Page 47
- > **In Memoriam: Gregory R. Choppin 1927-2015** – Page 49

Chair, 2015

Program Chair, 2015

**DAVID E. HOBART**

123 Big Oak Lane

Santa Rosa Beach FL 32459

Phone: (505) 227-4728

[dhobart15@gmail.com](mailto:dhobart15@gmail.com)

Chair-Elect, 2015

Program Chair, 2016

**PAUL BENNY**

Washington State University

Department of Chemistry

Pullman, WA 99164

Phone: (509) 335-3858

[bennyp@wsu.edu](mailto:bennyp@wsu.edu)

Vice-Chair, 2015

Program Chair, 2017

**LÆTITIA DELMAU**

Oak Ridge National Laboratory

Nuclear Material Processing Group

Oak Ridge, TN 37830

Phone: (865) 576-2093

[delmaulh@ornl.gov](mailto:delmaulh@ornl.gov)

Secretary, 2014-2016

**LUTHER McDONALD**

110 S Central Campus Dr

Salt Lake City, UT 84112

Phone: (509) 595-1101

[luther.mcdonald@utah.edu](mailto:luther.mcdonald@utah.edu)

Treasurer, 2014-2016

**ALICE MURRAY**

Savannah River National Laboratory

Aiken, SC 29808

Phone: (803) 725-0440

[alice.murray@srnl.doe.gov](mailto:alice.murray@srnl.doe.gov)

Councilors

**GRAHAM F. PEASLEE**, 2015-2017

[peaslee@hope.edu](mailto:peaslee@hope.edu)

**SILVIA JURISSON**, 2014-2016

[jurissons@missouri.edu](mailto:jurissons@missouri.edu)

Alternate Councilor

**DAWN A. SHAUGHNESSY**, 2015-2017

Members-at-Large, Executive Committee

**SUZANNE LAPÍ**, 2015-2017

**RICHARD ROBERTS**, 2013-2015

# ACS NUCL Division 50th Anniversary: Introduction

David E. Hobart



*David E. Hobart graduated with his PhD in analytical chemistry from the University of Tennessee, Knoxville. He was a postdoc at Oak Ridge National Laboratory and later accepted a staff position at Los Alamos National Laboratory during which he served two years at DOE Headquarters, Washington D.C. He also worked at Lawrence Berkeley Lab. and later as a technical consultant to Sandia National Laboratories. Returning to Los Alamos, he later retired and is presently a guest scientist at Los Alamos, a courtesy faculty member at Florida State University and 2015 Chair of the ACS Division of Nuclear Chemistry and Technology. His focus areas are lanthanide and actinide chemistry, actinides in the environment and nuclear waste disposal.*

The ACS Division of Nuclear Chemistry and Technology was initiated in 1955 as a subdivision of the Division of Industrial and Engineering Chemistry. Probationary divisional status was lifted in 1965. The Division's first symposium was held in Denver in 1964 and it is fitting that we kicked-off the 50th Anniversary in Denver in the spring of 2015. Listed as a small ACS Division with only about 1,000 members, NUCL's impact over the past fifty years has been remarkable. National ACS meetings have had many

symposia sponsored or co-sponsored by NUCL that included Nobel Laureates, U.S. Senators, other high-ranking officials and many students as speakers. The range of subjects has been exceptional as are the various prestigious awards established by the Division. Of major impact has been the past 30 years of the NUCL Nuclear Chemistry Summer Schools to help fill the void of qualified nuclear scientists and technicians. In celebrating the 50th Anniversary we honor the past, celebrate the present and shape the future of the Division and nuclear science and technology. To celebrate this auspicious occasion a commemorative lapel pin has been designed for distribution to NUCL Division members.

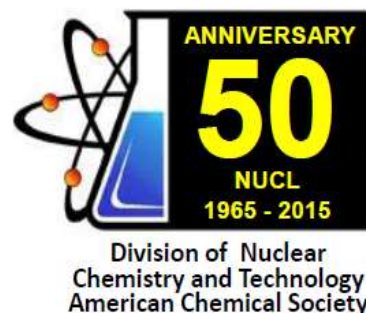


Figure 1. Anniversary lapel pin.

Formal steps were initiated in 1962 by Subdivision chair Joseph Martin. Morton Smutz was the first chair of the probationary division with support from Bernice Paige. William Morris and Clark Ice organized the first symposium in Denver in 1964. J. L. Schwenneson chaired the sessions. This symposium entitled, "Production Technology of Np-237 and Pu-238," broke new ground in presenting previously classified technology to the public. The Division of Nuclear Chemistry and Technology was originally designated as DNCT but the abbreviation was later changed to NUCL. In the succeeding fifty years, the range of topics presented at national ACS meetings has been extensive, including

fundamental nuclear research, creating new elements, nuclear physics, radiochemistry, analytical chemistry, environmental concerns, nuclear waste disposal, medical radioisotope production, nuclear fuels, etc. The longest-running symposium is Analytical Chemistry in Nuclear Technology which has been featured nearly annually for more than a decade. Most recently at the 2015 ACS National Meeting in Denver, the symposia convened were: The NUCL 50th Anniversary Symposium; The Seaborg Award Symposium in Honor of Heino Nitsche; Nuclear Forensics; and The Convergence of Theory and Experiment in Heavy Element Chemistry.

The many awards sponsored by NUCL over the years have included: The Glenn T. Seaborg Award in Nuclear Chemistry to recognize and encourage research in nuclear and radiochemistry or their applications; The W. Frank Kinard Distinguished Service Award recognizes NUCL members for outstanding service to the Division and the field of nuclear science; The Charles D. Coryell Award honoring undergraduate students who have completed research projects in nuclear or nuclear-related areas; and the Nuclear Summer Schools Outstanding Students Award.



Figure 2. Brookhaven Nuclear Summer School Outstanding Student Award recipient Aaron French as presented by J. David Robertson, ACS NUCL Division Summer School Director.

Of significant impact in forging the next generation of nuclear scientists is the ACS NUCL Undergraduate Summer Schools. The

first Nuclear Chemistry Summer School intended to educate students and help fill the void of qualified nuclear scientists and technicians was in 1984 at San Jose State University, CA. The subsequent 30 years of the Summer Schools has been highly effective and very successful by producing outstanding graduates. Present fellowships include a stipend of \$4000, all tuition and fees, transportation to and from the Summer School location, housing, books, and laboratory supplies. Transferable college credit will be awarded through the ACS accredited chemistry programs at San Jose State University (7 units) or the State University of New York at Stony Brook (6 units). The course will consist of lectures and laboratory work including introduction to state-of-the-art instrumentation. In addition to the formal instruction, the course will include a Guest Lecture Series and tours of nearby research centers at universities and national laboratories. Participants in the 2015 Summer Schools will be encouraged to join a research project during the following summer at a university or federal research institution. An "Outstanding Student" is selected from each summer school site. These students will be invited to attend the following spring national meeting of the American Chemical Society with all expenses paid.

Over the past five decades communications in the NUCL Division has tracked with the advance of technology. At all ACS national meetings the Division holds an Executive Committee Meeting and a Business Meeting followed by a Social Hour. The NUCL Division Newsletter is distributed quarterly by postal service and e-mail to all members that includes updates from the Division Chair, Program Chair, Treasurer, Summer School Director, and Award Nominations Committee.

The Newsletter contains information on upcoming meetings, abstract deadlines, job openings, etc. More recently, the Division has maintained an Internet web site with extensive information about NUCL activities: <http://www.nucl-ac.s.org/>

In the last fifty years NUCL Division members have authored countless journal articles, book chapters, books and popular press articles. Important book publications to name a few examples include: *The Chemistry of the Actinide Elements*, Second Edition (Katz, Seaborg and Morss, Eds.); *The Chemistry of the Actinide and Transactinide Elements*, Third Edition (Morss, Fuger and Edelstein, Eds.); *The Chemical Thermodynamics of Neptunium and Plutonium* and *The Chemical Thermodynamics of Americium* (OECD Nuclear Energy Agency); *Modern Nuclear Chemistry* (Loveland, Morrissey and Seaborg); *Radiochemistry and Nuclear Chemistry* (Choppin, et al.); and *Advances in Plutonium Chemistry 1967-2000* (Hoffman, Ed.). In progress is the rewrite of the classic text, *The Plutonium Handbook* (Clark, Geeson and Hanrahan, Eds.) dedicated to the 75th Anniversary of the discovery of plutonium by Seaborg and co-workers.

During the 50th Anniversary year it is useful to reflect on the many nuclear-related activities and incidents that have involved or affected members of the NUCL Division and the world at-large over the last five decades. For more than fifty years we have been living under the nuclear double-edged sword that offers the threat of devastating weapons of mass-destruction or the promise of unlimited power for future generations. There is no doubt that the Cuban Missile Crisis of 1961 was very close to initiating a nuclear world war. Calm political negotiations brought us back from the brink. The collapse of the Soviet Union in 1991 also lowered the nuclear crisis

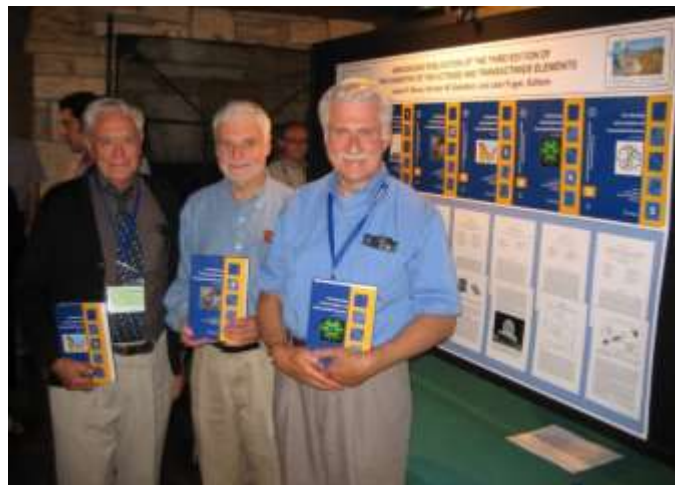


Figure 3. Editors of the *Chemistry of the Actinide and Transactinide Elements*. Many NUCL Division authors are major contributors to this classic monograph.

threshold, ushering in the end of the Cold War. With the end of the Cold War the United States had to change its nuclear weapons posture that ushered in the end of nuclear testing. No testing meant that we would then rely on science-based stockpile stewardship and computer simulation of weapons testing experiments. Treaties with the Russian Republic and other nations required a nuclear weapons stockpile reduction. With the lack of controlling and securing of special nuclear materials (SNM) in the Former Soviet Union, the US, other nations and the International Atomic Energy Agency (IAEA) stepped up to secure these locations and safeguard the SNM from illicit activities and potential diversion. Many nuclear scientists from the US and abroad supported the IAEA in controlling the abandoned nuclear facilities and aided in accounting for SNM. Closer to home and more recently, the closure of the Rocky Flats Plant north of Denver, CO meant that Los Alamos National Laboratory would assume a major role in weapons production capability.

The threat of nuclear terrorism has become a major concern for nations world-wide. The US and other countries have established agencies and programs aimed at nuclear materials proliferation prevention. For this reason nuclear forensics has become an important



science and a critical part of law enforcement. When illicit nuclear material is interdicted or suspected, law enforcement agencies need to answer very quickly the attribution questions: who, what, when, where, how, how much and why. Fission products uniquely identify a specific spent-fuel assembly for international safeguards to prevent and deter potential diversion and help to identify covert nuclear weapons programs. The nuclear fuel assembly power history is a distinct fingerprint and serves as the basis of a method for unique identification of attribution. Using fission product concentrations to characterize attribution seriously limits the ability of a proliferator to deceive law enforcement agencies.

For the first time in decades, less than 100 nuclear power reactors are operating in the US that are still producing 70% of the nation's electricity. Among the causes for this situation are safety concerns brought about by the 1979 Three Mile Island, 1986 Chernobyl, and 2011 Fukushima Daiichi reactor incidents. Furthermore, relatively inexpensive natural gas and oil prices and policies favoring renewable power resources such as wind, solar, etc. have slowed the nuclear power industry. One of the most pressing issues presently impeding the renaissance of nuclear power generation in the US is that of safe nuclear waste disposal. The problem of nuclear waste disposal is urgent. Leaving the radioactive waste issue unsolved creates permanent and tempting targets for terrorist and threatens our health and the environment. We have a moral imperative to solve this problem so we do not burden our children, grandchildren and untold future generations.

Present US Nuclear Regulatory Commission policy separates nuclear waste into two main categories. The first category is civilian nuclear waste generated as a by-product of electrical power generation. This consists

mainly of high-level waste that is primarily spent nuclear fuel rods and fission products. The second category is defense nuclear waste resulting from national defense activities and this consists mainly of low-level waste including light actinides, fission products etc. Other minor miscellaneous waste categories include smoke detector Am-243, medical radioisotopes waste, X-ray, gamma ray and neutron sources for drilling exploration, industrial-scale metal production and plutonium-238 waste from space exploration programs.

The Yucca Mountain site adjacent to the former Nevada Nuclear Test Site was the proposed site for civilian nuclear waste. The isolated location above the water table and in welded volcanic ash (tuff) the Yucca Mountain Site was thought to be adequate for containing nuclear waste for a million years. Recently however, concerns over potential volcanism and sooner than anticipated surface water infiltration prompted the Obama administration to cancel the project. This leaves the disposition of civilian nuclear waste in limbo at present. The disposition of defense nuclear waste has been addressed by the Waste Isolation Pilot Plant (WIPP) in southeastern New Mexico near Carlsbad. WIPP is a licensed permanent nuclear waste repository. A half-mile deep in a bedded salt formation was chosen because of its remote location, easy mining operations, long-term geologic stability and self-sealing properties. WIPP now holds more than 171,000 waste containers containing about 4.9 metric tons of plutonium and other nuclear waste elements. Aside from recent safety issues at the WIPP that resulted in temporary closure, it remains one of the best options for nuclear waste disposition. There has been recent discussion that a WIPP-II for civilian high-level waste may be a viable option as well.

Significant achievements have been made in the last 50 years in heavy element science.



Figure 4. Waste Isolation Pilot Plant near Carlsbad, NM.

Mendelevium, element 101, was discovered by Seaborg and co-workers at Berkeley Laboratory in 1955 and in 1958 nobelium, element 102, was discovered at Berkeley. It was after much controversy and conflicting claims from various institutes that the first complete and incontrovertible report of its detection only came in 1966 from the Joint Institute of Nuclear Research at Dubna in the former Soviet Union. Earlier, in January 1958, the U.S. Atomic Energy Commission reviewed the status of transuranium isotope production in the US and built the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory with a fundamental focus on isotope research and production. Having gone critical in 1965, the in-core uses for HFIR have since broadened to include materials research, fuels research, fusion energy research, isotope production and research for medical, nuclear, detector and safety purposes. Additionally, the HFIR is providing targets for nuclear bombardment reactions to countries around the world in the quest to discover new heavy elements. In the last fifty years 17 new elements have been discovered from Atomic Number 101 to 118, with only 4 remaining unnamed. The discovery of new elements has been a multi-national endeavor with many researchers working on the projects and involving multiple nuclear facilities. At the forefront of new element discoveries are Lawrence Berkeley National Laboratory, Lawrence Livermore National Laboratory,

GSI Helmholtz Centre for Heavy Ion Research, Darmstadt, Germany, and the Flerov Laboratory Joint Institute for Nuclear Research, Dubna, Russia. Ongoing efforts to discover element 119 and 120 and the elusive “Island of Stability” are underway at present in a collaboration between these laboratories.

For more than five decades, radioisotope thermoelectric generators (RTGs) have played a crucial role in the exploration of space, enabling missions of scientific discovery to destinations across the solar system. Strontium-90, americium-241 and plutonium-238 are commonly used in RTGs with Pu-238 the most desirable power system. RTGs are useful where power from solar radiation is not a viable option because of large distances from the Sun. An RTG power system with no moving parts transforms the heat from alpha or beta decay directly into electricity using solid-state thermoelectric converters, which generate electricity using the flow of heat from the large temperature difference between the hot nuclear fuel and the cold space environment. Advances in computer controlled robotics have made long missions possible without jeopardizing human life. Pu-238 also keeps the electronics and computer systems operationally warm in the cold depths of space. These voyages revealed the nature of Earth's moon, icy geysers and sulfur volcanoes on moons of Saturn and Jupiter, and sustained long journeys to the outer reaches of our solar system. The Voyager 1, for example, has passed out of the Solar System. Voyager 1 was launched by NASA in September 1977 to study the outer Solar System. Operating for over 37 years, the spacecraft still communicates with the Deep Space Network to receive routine commands and return data. At a distance of about 130.62 AU ( $1.954 \times 10^{10}$  to  $10^{11}$  km), it is the farthest spacecraft from Earth. Voyager 1 has three radioisotope thermoelectric generators (RTGs) mounted on a boom. The RTGs generated about 470 watts of electric power at the time of launch, with

the remainder being dissipated as waste heat. The power output of the RTGs does decline over time (due to the short 87.7-year half-life of the Pu-238 and degradation of the thermocouples), but the RTGs of Voyager 1 will continue to support some of its operations until 2025.

Cassini–Huygens is an unmanned spacecraft sent to the planet Saturn. Cassini has studied the planet and its many natural satellites since arriving there in 2004. Developed starting in the 1980s, the design includes a Saturn orbiter, and a lander for the moon Titan. The Huygens craft landed on Titan in 2005. The two-part spacecraft is named after astronomers Giovanni Cassini and Christiaan Huygens. The Cassini orbiter is powered by three radioisotope thermoelectric generators (RTGs), which use heat from the natural decay of about 33 kg (73 lbs.) of plutonium-238 as the dioxide to generate direct current electricity via thermoelectrics. The RTGs were designed to have very long operational lifetimes.

The Mars Science Laboratory Mission Rover named “Curiosity” is also powered by radioisotope power generators. This power source gives the mission an operating lifespan on Mars' surface of at least a full Martian year (687 Earth days) or more while also providing significantly greater mobility and operational flexibility, enhanced science payload capability, and exploration of a much larger range of latitudes and altitudes than was possible on previous missions to Mars.

The most recent NASA mission launched in January 2015 is the exploration of the planetoid Pluto, which is the origin of the name of the element plutonium named by Glenn Seaborg. Electrical power for the New Horizons is furnished by a single RTG. The compact, rugged General Purpose Heat Source (GPHS)-RTG aboard New Horizons, developed and provided by the U.S.

Department of Energy, carries approximately 11 kilograms (24 pounds) of plutonium dioxide fuel. The New Horizons spacecraft was designed to make the first close-up study of Pluto and its moons and other icy worlds in the distant Kuiper Belt. The spacecraft has seven scientific instruments to study the atmospheres, surfaces, interiors and intriguing environments of Pluto and its distant neighbors.



Figure 5. Artist's rendering of the New Horizons during its Pluto flyby. The RTG assembly is on the boom at lower left.

Of major impact in the last few decades is the advent of the Information Age and the power of the Internet for instantly accessing chemical information (or any information for that matter). With a virtual library subscription, users can access nearly any book or journal article that has been made available digitally. Web sites like Wikipedia offer rapid access to a variety of science topics. Additionally, users can contribute their own knowledge to the information pool.

In conclusion, the ACS-NUCL Divisions impact has been remarkable over the last 50 years and it is expected that the Division will continue to serve its members, science and society over the next 50 years. Happy Anniversary NUCL!

# From the Cold War through Nuclear Deterrence: Four Decades at the Savannah River Laboratory

Don Orth



*Don was born in San Francisco, California in 1923. He obtained his B.S. degree in chemistry from the University of California at Berkeley in 1948 and his Ph.D. in chemistry also at UC Berkeley working with Glenn Seaborg in 1951. Don worked at the Savannah River Plant and the Savannah River Laboratory (now Savannah River National Laboratory) from 1952 until his retirement in 1992. In 1990 Don was awarded the Glenn T. Seaborg Award in Actinide Separations. In 1993 the Savannah River Laboratory created its highest award, the Don Orth Award, to honor scientists for technical excellence and service to the Savannah River Site.*

I was born and raised in San Francisco and graduated as valedictorian of San Mateo High School in 1941. I entered the University of California at Berkeley in the fall of 1941 and decided to study chemistry since I particularly enjoyed my high school chemistry classes. During my first semester at UC Berkeley, I was having a seaside picnic with my family on a pleasant Sunday south of San Francisco when I learned of an event that would alter my life for a few years. A guy walking along the shore yelled to us that Pearl Harbor had

been attacked and that we were at war. I was able to complete one more semester of school but then was drafted into the U.S. Army in January, 1943. The better part of the next two years I spent in training: medic; engineer; chemical warfare; signal corps; and intensive chemical engineering courses under the Army Specialized Training Program.

In the fall of 1944, my family and I learned the tragic news that my older brother Dick had been killed in the Battle of Leyte Gulf in the Philippines. His submarine was sunk near the end of the battle and all the men on board were lost.

In the winter of 1944, those of us who considered ourselves to be perpetual trainees were assembled in Europe to prepare to cross the Rhine and defeat the German armies. Given the first service choice I volunteered for the 17th Airborne Division glider troops. However, my training instead resulted in my assignment to the 17th Airborne Division signal company. The crossing went so well that I crossed the Rhine over a pontoon bridge in the back of a truck.

As action in Europe ceased, those of us with minimal combat action were to be shipped to the Pacific theatre to prepare for the invasion of Japan. As we left the Mediterranean Sea, we were told that a new weapon had been used and that the war was over. Our joy was unrestrained when the ship captain told us that our new destination would be Newport News, Virginia for eventual discharge from the service.

I went back to Berkeley in February of 1946 and finished my B.S. degree in 1948. I joined Glenn Seaborg's research group at Berkeley to work on my Ph.D. These were extremely



exciting times to be doing research in nuclear chemistry and even that may be something of an understatement. Glenn's graduate students worked closely with Al Ghiorso to design experiments using the cyclotron. Nothing in my career ever quite matched the fun of doing chemistry on the cyclotron targets with the potential of discovering new elements and new and interesting isotopes. One part of my graduate school career that I well remember is Glenn's Thursday morning group meetings that were informal and open for students to discuss research activities. Although Glenn never said explicitly that you had to discuss your research each week, I noticed that those who did not say much were soon no longer part of the research group.

The E.I. DuPont Company hired me after I obtained my Ph.D. in January, 1951 to join the staff of scientists and engineers to design and operate the Savannah River Plant near Aiken, South Carolina. When I was hired the plant and laboratories were being constructed and so I was sent first to Argonne and then to Oak Ridge where we performed shielded cell pilot scale tests of the newly developed PUREX process. During this period I met my future wife, Knoxville, Tennessee native Jean Ledgerwood. I asked my friend and fellow UC Berkeley graduate Dave Karraker to be my Best Man at the wedding. Jean seemed to know just about everyone in East Tennessee so she had plenty of family and friends at the wedding. I had discussed with Jean before we were married if she would be willing to move to South Carolina to be there at the beginning of the huge new Savannah River Plant. She agreed and we recently celebrated our 62nd wedding anniversary.

The Savannah River Plant was designed to be the sister plant of the Hanford Plant in Washington to produce plutonium and tritium for the nuclear weapons program. E.I. DuPont at Hanford had used the bismuth phosphate precipitation process in the route to plutonium

metal. However, it was clear to many of us that using solvent extraction with tributyl phosphate in the PUREX process was superior to bismuth phosphate precipitation. This put us at odds with some DuPont personnel who wanted to use the same process that they were familiar with at Hanford. When we decided on PUREX at Savannah River, some DuPont personnel retired rather than be on the design and implementation team.

My four decades of contributions (along with the contributions of many thousands of others) to the Separations Department at Savannah River helped keep the Cold War cold through nuclear deterrence. However, the fact that Savannah River would eventually have five nuclear reactors and that nuclear chemistry was still relatively new led to my participation in many interesting nuclear experiments outside the realm of process development and support. Performing basic research in nuclear chemistry and radiochemistry required researchers to stay abreast of this rapidly expanding field. Formation of the ACS Division of Nuclear Chemistry and Technology in the mid-1960's encouraged professional interactions and information exchange. I was active in the NUCL for many years and served on the Executive Committee from 1967-69 and again from 1976-78.

I join thousands of others in congratulating NUCL for achieving 50 years of outstanding service to nuclear scientists throughout the world. If I were a bit younger I would have loved to be in Denver to join the celebration! I also want to say to young folks reading this NUCL 50th anniversary commemorative booklet that my 50 years in nuclear science were wonderful and I could not have chosen a more challenging and rewarding career.

# Reflections on My 70 Years in Chemistry

Darleane Christian Hoffman



*Darleane was born in Terril, Iowa, November 8, 1926. She received her B. S. (1948) & Ph. D. (1951) degrees from Iowa State College, Ames, Iowa. Oak Ridge National Laboratory: Chemist, 1952-53; Los Alamos Scientific Laboratory: Staff Member, Project Leader, Assoc. Gp. Leader: 1953-78; Los Alamos National Laboratory, Division Leader Chemistry-Nuclear Chemistry, 1979-82; Division Leader, Isotope and Nuclear Chemistry 1982-84. Tenured Prof. Chem., University of California, Berkeley 1984-91; Professor Emerita 1991-present; Faculty Sr. Scientist, Nuclear Science Division, Lawrence Berkeley National Laboratory (LBNL) 1984-present. Although officially retired, she still serves as Advisor for graduate students and as a member of Ph. D. qualifying exam committees and as reader on dissertations. She is a Fellow of Norwegian Academy of Science and Letters; American Association for the Advancement of Science; American Institute of Chemists; American Physical Society; American Academy of Arts and Sciences. Major awards include: Honorary Doctorates: Clark University, USA (2000); University of Bern, Switzerland (2001); NSF Sr.*

*Postdoctoral Fellowship, Institute for Atomic Energy, Kjeller, Norway, 1964-65; Guggenheim Fellowship, LBNL, 1978-79; American Chemical Society (ACS) Award for Nuclear Chemistry, 1983 (first woman recipient); ACS Garvan-Olin Medal, 1990; U. S. National Medal of Science, 1997; ACS Priestley Medal, 2000; Sigma Xi Proctor Prize for Scientific Achievement, 2003; John V. Atanasoff Research and Discovery Award, Iowa State University, 2007; Hevesy Medal Award 2010, and the 2014 Los Alamos National Laboratory Medal.*

It was a wonderful experience for me to be able to attend and participate in the March 24-25, 2015 Symposium celebrating the 50th Anniversary of the ACS Division of Chemistry and Technology (NUCL) in Denver. This Celebration prompted me to recall some of my more memorable experiences specifically related to the NUCL Division as well as some from my long career in nuclear and radiochemistry and consider what I was doing 50 years ago!

After living in Los Alamos where my husband Marvin and I had worked since 1953 at the Los Alamos Scientific Laboratory, in the Anniversary year of 1965 our family was preparing to spend a sabbatical in Oslo, Norway. We now had 2 children (Maureane, born 1957 and Daryl, born 1959) and, fortunately, my mother who had been living near us and taking care of the children since I worked full-time plus many evenings and weekends as well, was able to accompany us. Marvin was awarded a Fulbright Fellowship for research at the University of Oslo, Norway and I received an NSF Postdoctoral Fellowship which I could take to most any suitable institution. This worked out very well since I was happy to spend the year in Norway as my father's family had emigrated

to the U. S. from Norway in the 1840's and I wanted to visit the places they came from. In addition, I had some scientific contacts there. We had a most rewarding experience, both personally and scientifically. We made many new friends and interacted with colleagues with whom we still communicate. Marvin conducted research at the Van de Graaff Facility at the University of Oslo and I studied properties of short-lived fission products at the Institute for Atomic Energy in Kjeller, Norway, a short distance from Oslo.

Later, in 1978-79, we spent a sabbatical year in Berkeley where I was associated with Professor Glenn T. Seaborg's Heavy Element Nuclear and Radiochemistry group at Lawrence Berkeley Laboratory and attended his Monday noon 'sack lunch' group meetings and listened to brief research progress reports from group members. I was also privileged to meet Albert Ghiorso who helped build the apparatus at the 88-Inch Cyclotron that I and co-workers used to study short-lived isotopes of heavy element isotopes. This sabbatical year was later to have a tremendous impact on my future career! I left Berkeley in August 1979 (a little early) to return to Los Alamos to become Leader of the Chemistry-Nuclear Chemistry Division, the first woman leader of a technical division at Los Alamos Scientific Laboratory (LASL).

One of my early and especially memorable occasions was receiving the Award in Nuclear Chemistry (now known as the Seaborg Award) in 1983 as the first woman recipient. It was presented to me in March 1983 at the ACS National Meeting in Seattle by Prof. Seaborg (ACS 1976 Past-President). A very nice color portrait of the photo of me in my red dress hung in his office at LBL for several years until he replaced it with a photo of himself with the movie star Ann-Margret, (Swedish) I think! I am half Norwegian and Glenn was very proud of his Swedish heritage. Swedes and Norwegians have a long history of not

getting along with each other so we used to make jokes about that! Anyway, I blamed it on the fact that she was Swedish, never mind she was also a movie star!



Figure 1. Glenn Seaborg presents the ACS award in Nuclear Chemistry to Darleane Hoffman, March 1983. The next woman to receive this award was Joanna Fowler in 2002, followed by Silvia Jurisson in 2012.

In August 1989, a "Confidential" letter from the ACS informed Joe Peterson, then Program Chair of NUCL, of Darleane's receipt of the Garvan Medal which was to be presented on April 28th at the Awards Banquet during the 199th ACS National meeting in Boston, April, 1990. It asked if he could find a place in the NUCL program for my Award Address and to contact me directly and in confidence about this until after it was announced in Sept. 1989 at the ACS National Meeting in Miami. Previously, Joe had also talked me into helping Dave O'Kelley as a co-organizer of the overall program for our 25th Anniversary Celebration. When we subsequently learned that I was to receive the Garvan Medal, I received a call from Joe who graciously offered to organize another special symposium for this award, but knowing how difficult that would be and all the work that Gerhart and Ellis had done to organize this Symposium I felt it would be most appropriate for me to give my Garvan address in the NUCL program. After all much of my "service to chemistry" (if any) had been linked to NUCL and most of my research was reported

in its symposia. Although not quite a Charter Member of NUCL, I have ‘grown up’ or ‘grown old’ with NUCL and felt very strongly about the importance of educating young scientists in nuclear and radiochemistry and its myriad of applications.

Joe Peterson, did, indeed, find a place for me in the 1990 NUCL program: 40 minutes + 10 minutes for Discussion. It was a special honor for me to give the Garvan Medal Address in the 1990 NUCL program, the 25th Anniversary of our NUCL Division. Although the Garvan Medal is only awarded to women, it includes women from all disciplines of chemistry and, consequently, the competition is very strong.

Garvan Medal Citation: *“The 1990 Garvan Medal is presented to Darleane C. Hoffman for her many fundamental contributions to the physics and chemistry of the heaviest elements and for her leadership and service in the area of nuclear and radiochemistry.”*

I regarded my selection for the Garvan Medal as recognition of the importance of nuclear and radiochemistry in modern society. I chose the title “The Heaviest Elements: Past (1789-1989), Present (1990), and Future

(2015)”, specifically for the 25th Anniversary of our NUCL Division. (I pointed out that the term “Heaviest Elements” is a ‘moving target’). I included my “Cloudy Crystal Ball” predictions for 2015, or 25 years into the future.

Present: 1990 Status

- Chemical Studies: None performed beyond Element 105.
- Nuclear Studies: Periodic Table showed 108 elements.

My “Cloudy Crystal Ball” predictions for 2015 included:

- Discovery of 6-10 New Elements
- Production of new neutron-rich isotopes of odd-Z with longer half-lives.
- Use of ‘atom-at-a-time’ techniques for chemical & nuclear studies.
- Chemical studies to search for “relativistic” effects which are expected to increase as  $Z^2$  and may cause deviations from expected trends will be investigated.
- New information on systematics of spontaneous and neutron-induced fission will spark a “renaissance” of interest in fission phenomena and its role in production of new isotopes and elements



# Challenges and Rewards in Nuclear Chemistry

Paul Karol



*Paul Karol was born in the Bronx in 1941. He graduated from Eastchester (NY) High School in 1957. His B. A. in chemistry was from Johns Hopkins University in 1961 and his Ph. D. in nuclear chemistry was from Columbia University under the auspices of Jack Miller. Paul was a post-doctoral researcher with Gerhart Friedlander for two years at Brookhaven National Laboratory studying high energy fission and on the faculty at Carnegie Mellon University for over forty years. He served for five years as Associate Dean of Science and retired in 2012 as Professor Emeritus of Chemistry. He was Chair of the ACS Division of Nuclear Chemistry and Technology in 1996 and of the ACS Committee on Nomenclature where he has continued to be active. Paul was Chair of the IUPAC Commission on Nuclear and Radioanalytical Techniques and has been four-time Chair of the IUPAC/IUPAP Joint Working Party on the discovery of new elements. He is now living in Palo Alto near his grandchildren and working on a number of books. In his non-academic life, Paul played lacrosse for the New York Lacrosse Club and also the Pittsburgh Lacrosse Club and takes pleasure in butchering classical music on his piano.*

**Birth:** Applications to college asked for one's intended major. I had no idea where I was going. I had done well in high school

chemistry, so I entered "chemistry" as my major since a blank entry was not acceptable. In my freshman year as a chemistry major at Johns Hopkins, we were using as a text Linus Pauling's College Chemistry and at the last lecture, the professor apologized for not getting to the final chapter which was on "nuclear chemistry". I had never heard of nuclear chemistry before, had no idea what it was, but was struck by how cool it sounded. So I decided that was the field of chemistry I was going to pursue. Many times over the years as a professor myself, I have told this vignette to my students and said that was the worst conceivable way to make a life-long decision, but I turned out to be lucky. And so it started.

In my junior year I got permission to enroll in the graduate course "Nuclear Chemistry" where the text was Friedlander and Kennedy's classic. There were only two other students enrolled, both much older graduate students. I aced it! My senior year I wrote to Friedlander and asked for recommendations on graduate schools in nuclear chemistry. The choice I made was Columbia with Jack Miller.

**Youth:** My research at Columbia focused on high-energy spallation of medium mass nuclei using radiochemical separations, beta proportional counters and sodium iodide counters with newly available 100-channel multichannel analyzers. "High-energy" in those days was the 370 MeV proton beam from a synchrocyclotron. The bombardments were done in a suburb of New York City at Columbia's Nevis Laboratories, originally part of Alexander Hamilton's estate (he of \$10 bill fame and named after the island Nevis in the Caribbean where he was born). I actually had to set up a wet lab in what was Alexander Hamilton's stable in order to do my radiochem

separations as soon after bombardment as possible.

My research evolved into assessing the role of the nuclear surface on yields of neutron knockout reactions (and in recent years has broadened to interest in halo nuclei studies). When my preliminary results proved very interesting, Jack Miller had me present them at the Division of Nuclear Chemistry and Technology session in the Detroit ACS meeting in 1965, fifty years ago. I don't remember too much about my performance other than thinking "Who am I? Why am I here?", words that became famous later during a presidential election. I'm sure I was catatonic during my brief solo.

**Adulthood:** After getting my degree from Columbia, I went to work with Gerhart Friedlander at Brookhaven National Laboratory studying high-energy fission. By now, "high energy" was 28 GeV. My particular attention was to the production of the heavy rare earth products thus launching my new interests in rare earth chemistry and ion-exchange separations both of which were added to my adult repertoire in chemistry. Opportunities to expand one's exposure to science were nearly overwhelming at Brookhaven. One of the talks I went to was on "positronium chemistry" by V. Goldanskii from Russia. I was hooked on the esoteric nature of these studies and they would occupy me for a number of years subsequently.

**Middle Age:** In 1969, I joined the faculty at Carnegie Mellon University which had recently been formed by the merger of the Carnegie Institute of Technology and the Mellon Institute (a research center) and was expanding its chemistry department. I got involved in a fascinating collaboration with Truman Kohman on analyzing lunar dust returned by the Apollo missions to enable Nd-Sm dating of lunar samples through isochron isotopic studies. My expertise in clean

separations of neodymium and samarium were a synergistic contribution.

I also launched a many-year series of experiments on general trends in high-energy spallation using alpha beams at the Space Radiation Effects Laboratory accelerator facility in Virginia and a few years later by employing protons and pi mesons at the Los Alamos Meson Production Facility.

My seduction into positronium studies led to some doctoral theses and involved interesting instrumentation challenges but also to my co-chairing and organizing an ACS Symposium on Positronium Studies in Chemistry with Mike Fluss, now at Livermore National Laboratory.

The National Research Council of the National Academy of Sciences had a Commission on Nuclear and Radiochemistry, chaired by Greg Choppin, which I was recruited to join. A major topic was the national need for nuclear and radiochemists and the fact that this need was not being filled by a sufficient influx of students. A workshop and subsequent report was generated in 1978 in which the argument was made that it behooved the government to establish a summer school in nuclear and radiochemistry as a means to address the manpower issue. Besides myself, I remember the Committee members included Darleane Hoffman, Trish Baisden, Gerhart Friedlander, Rolf Herber and Ed Macias among others. The history of that proposal is the extraordinary successful implementation of the Nuclear Chemistry Summer School at San Jose and Brookhaven summer programs.

I have had the pleasure of presenting lectures at both the San Jose and Brookhaven summer schools over the intervening decades, usually on nuclear structure and reactions, and also guest lectures on my now fifty-year old interest "High Energy Nuclear Reactions: From the nuclear surface to beyond the

galaxy's edge". The latter twist grew from my interest in ultrarelativistic cosmic rays.

In 1994 I was fortunate to be seated with Darleane Hoffman and Glenn Seaborg at the 1994 ACS National Meeting in San Diego when the announcement was made confirming the discovery of element 106 and proposing it be named seaborgium. That announcement sprouted an eventful distraction for me, the protocol for naming newly discovered elements. There were already long-standing debates about priorities for elements 104 and 105. To this was now added the perceived audacity of naming an element, 106, after a living person. The perception was on the part of some subtly anti-American members of the heavy element "kinship" (sic). I enlisted as a tactical combatant in what I was to soon refer to in public as the "Transferrmium Wars". The Bronx genomes in my DNA enabled me to write an 8-page white paper ridiculing IUPAC for its hypocrisy in interfering with the historic tradition element naming protocol. I sent the diatribe to forty national adhering organizations of IUPAC, soliciting their support, and got the ACS Committee on Nomenclature to convince the ACS Executive Bureau to object to IUPAC's process. IUPAC was put in the position of having to rescind their previous misguided decision and finally sanction seaborgium.

In 1996, I got together with my mentor Gerhart Friedlander and we organized and co-chaired an ACS symposium in New Orleans to celebrate the 100th anniversary of the discovery of radioactivity which got some very nice exposure in the press. The coterie of presenters was very international and distinguished as can be seen in the photo below, published in Chemical & Engineering News a bit after the meeting.

**Dotage:** Partially to punish me for interfering with IUPAC's role in the



Figure 1. Participants at the symposium on "Centennial of the Discovery of Radioactivity" included: (front row, from left) Alfred P. Wolf, Guenter Herrmann, R.J. Silva, Glenn T. Seaborg, Jean J. Fuger, Robert Guillaumont, Jean-Pierre Adloff, Leonard W. Fine, and Joseph Cerny; and back row (from left) Hans R. von Gunten, Petr Benes, Heino Nitsche, Steven W. Yates, Gerhart Friedlander, Joseph R. Peterson, and Paul J. Karol.

Transferrmium Wars, I was appointed Chair of a newly formed "Joint Working Party" on the discovery of new elements. The turn of the millennium has seen us address claims for nine transmeitnerium elements that hit the world stage. By the time this memoir is in press, all those claims should have formal recommendations about their legitimacy in terms of having satisfied (or not) accepted criteria for discovery.



Figure 2. Photograph of (from left) Meryl, Barbara, Gerhart, and Paul.

Brookhaven National Laboratory celebrated the 90th birthday of Gerhart Friedlander in 2006 and I was thrilled to reminisce about our

years of friendship, collaboration and science together. Shown in Figure 2, from left to right, are my wife (Meryl), Gerhart's wife (Barbara), Gerhart Friedlander, and yours truly at that Friedlanderfest.

In retrospect, what do I appreciate most about my half century of nuclear chemiphilia? I

would say it's kept me feeling young and excited about watching and participating in continued advances in the field and meeting the upcoming generations of replacement troops.

But none from the Bronx! Sigh.



# Probing and Understanding the Science of the Actinide Elements

*Richard Haire*



*institutions as well as having them perform research studies at ORNL. One institution in this regard for over three decades was the Institute for Transuranium Elements (European Commission) in Germany. He has several hundred journal publications and is the author of a number of chapters in reference books on the f-elements. Dick has pursued the gas, solution, and solid state science of the actinides in multiple collaborate studies, and developed unique synthetic research approaches for the actinides from Ac through Md. He continues as a consultant involving multiple aspects of these elements.*

*Born and essentially raised in Chicago, IL, Dick did his undergraduate studies in Illinois and obtained his PhD in chemistry in Michigan. He served as an assistant professor for honor's chemistry for one year, and also served as an adjunct professor in Nuclear Science at the U. of Tenn. and had worked for a short period at Argonne National Laboratory. He became a research staff member at Oak Ridge National Laboratory when the High Flux Research reactor (HFIR) became operational there in 1965. He was attracted to ORNL by the upcoming availability of weighable quantities of transplutonium elements from HFIR. He subsequently became the Group Leader at the Transuranium Research Laboratory, which was dedicated to research of the physics and chemistry of actinide elements for over four decades. Dick became a Corporate Fellow at ORNL; in 2013 he was awarded the Glen T. Seaborg Award in Nuclear Science.*

While NUCL is celebrating its 50<sup>th</sup> Anniversary, interest in and studies of the actinide series of elements proceeded the year 1965. While the elements from Ac-U were known and studied previously, it was the discovery of the man-made transuranium elements and subsequently the transplutonium elements (early 1940's) that launched the scientific efforts for the actinides series (the 5f- electron series). Initially, differences between the Th-Pu elements behaviors with those of the lanthanides (4f electron series) caused some confusion in understanding the actinide series, but this was quickly resolved. It became understood that transplutonium elements would also have stable trivalent states. This history is well described in the book by Seaborg and Loveland, "The Elements Beyond Uranium" (Wiley & Sons, Inc., 1990), and this book is recommended reading. The elements from Am-Cf were found to be more comparable in their behaviors to the lanthanide elements, until the science of the Es-Lr elements came into focus.

Since the 50<sup>th</sup> Anniversary of NUCL refers back to 1965, the date for my arrival at ORNL, NUCL Newsletter, 50<sup>th</sup> Anniversary, Page - 17

*Dick has been a guest scientist at a number of universities and institutions around the world. He has had multiple collaborations with scientists and post docs from several different countries, performing research both in their*

the discussion here is limited to a short overview of my efforts at ORNL in pursuing the chemistry and physics of the actinides in many collaborations and with scientists around the globe. A very important driver for these actinide efforts was the HFIR, which began isotope production of transplutonium elements after Seaborg, as Chairman of the AEC, helped organized the establishment of HFIR at ORNL. The HFIR provided weighable quantities rather than just multiple atoms of the transplutonium elements for research. This resulted in a tremendous surge in research of these elements, and has generated a great deal of scientific knowledge involving their chemistry, physics and nuclear nature. At ORNL, I was fortunate to become involved in a large number of collaborations on these elements: these efforts involved being a guest scientist at multiple national laboratories, universities, etc. around the world, as well as hosting scientists and post docs at ORNL for performing collaborative studies. The institutions which were involved and the source of visiting scientists were located in the both United States and in many foreign countries. Examples of the latter are (in no specific order): Belgium; France; Portugal; Germany; Switzerland; Austria; Australia; Japan, Russia, the US and more. Two of the longer collaborative efforts were with the Institute of Transuranium Elements, Karlsruhe, Germany (ITU; over three decades); and the former Japan Atomic Energy Research Institute (JAERI; 5 years). The enthalpy of solution investigations of select transuranium metals were performed early both at the University Liege in Belgium and at ORNL with Professor J. Fuger. Multiple collaborative investigations were then done with scientists at ITU in Germany; one important topic here was the high-pressure behaviors of actinide metals from Am through californium and that of several transplutonium compounds, mainly with Dr. Benedict and others at ITU. Several of these

high-pressure studies also involved the European Synchrotron Radiation Facility (ESRF) in Grenoble, France. Multiple studies of the enthalpy of sublimation of transplutonium metals compounds were investigated at Los Alamos National Laboratory and at ORNL. Dr. J.K. Gibson (now at LBL) was involved in the enthalpy studies of Es and Fm metals at ORNL. The Japan efforts revolved around multiple studies of actinides through a program for which I was acting as the leader at ORNL. A special and important collaboration involved the association of ORNL with the U. of Tenn., USA, and the presence/involvement of Prof. J. R. Peterson and his students.

An important aspect of this actinide work involved the purification and syntheses of special materials. This is an important aspect for studies of the transplutonium elements and their compounds for multiple reasons. One cannot simply order a product from a commercial supplier – rather, given their limited quantities, radioactive natures, the purities demanded for these studies, etc. The syntheses became an important and integral part of the different research studies. A specific example here is that encountered with einsteinium. The main isotope of einsteinium used was Es-253 (half-life of 20.5 days); it is mainly an alpha emitter. But working with the Es-253 has its “problems and difficulties”: One being it has decay heat of ~ 1000 watts per gram; the other is it produces a very high emission of 6.6 MeV alpha particles. This together with the energetic recoiling nuclei from decay caused very significant damage in the samples, and allows only a short period of time for studies. In addition, given the rapid in growth of daughter products (impurities) after roughly two days following purification, one has only a brief time before significant amounts of the Bk daughters have grown back in. Thus efforts had to be performed on only microgram quantities of Es on a tight

schedule. Studies with Fm were even more difficult.

A significant part of my involvements also focused on preparations of the small, special research samples desired. While many times a challenge, this aspect was both necessary and satisfying, especially when successful and meaningful data are obtained. Many people often do not realize the importance of high-purity sample materials that can make or break “attempted research” and obtaining meaningful data. Too often, samples are accepted as initially “labeled”. While this can happen with many non-radioactive materials, it is a special problem with radioactive materials, as samples are transformed into other elements.

An extremely valuable aspect of successful actinide research is collaborative work. This is an important philosophy of having a strong team concept with individuals that are very interested in pursuing and developing a correct understanding of these elements’ science. A great number of long lasting friendships were developed in these collaborative investigations, and many of these have been maintained. It was an exciting, rewarding and fun period!

In looking back, I have been surprised regarding the number of collaborations I had been involved with; so many efforts with many different experts and the vast array of the scientific topics that were covered in these actinide studies. Interests covered the gas, liquid and solid state phases of the actinides, and examining both compounds and their metallic states to try to fully understand the role and the changes resulting from the influence of the 5f electrons.

Space here is limited to fully discuss what we accomplished during the Nuclear Division’s 50 Year Anniversary Period. Perhaps a bird’s eye view of the efforts and types of studies

done during this period can be generated simply by outlining techniques used for the various studies of the actinide metals and their compounds. Thus, some specific investigations involved: (1) electron microscopy and diffraction; (2) photoelectron microscopy and determining electron energies for the actinides from Ac through Es; (3) determining the enthalpy of solution for the Am-Cf metals; determining the enthalpy of sublimation of the Am-Fm metals; high-pressure studies of the actinide metals, and the high-pressure phase behaviors and bonding of actinide compounds; (4) multiple types of absorption spectroscopy (phase behaviors, oxidation state other changes), (5) magnetic susceptibility investigations of the metals and compounds; and so on. For these studies, many pieces of special equipment had to be designed, acquired, set up and made available for the radioactive studies. Also, many special preparative techniques were developed and used for syntheses of the special forms of samples used in the multiple studies during this 50 Year Period.

One of the important aspects sought during this anniversary period is to understand the nature and science of these transuranium elements, especially the role of their electronic configurations and the differences in behavior between them and the 4f electron “sister” series. These differences between the 4f and 5f electron elements are summarized in Figure 1.

One immediate difference is observed for the early members of each series; the nature of the Th-Pu elements is notably different than that for the first 4f-series elements. As mentioned earlier, this factor initially misled early researchers’ thoughts about the two series.

### The Two f-Electron Series

LANTHANIDE METALS															
La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	
ACTINIDE METALS															
Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr	

Figure 1. Periodic table of the 4f and 5f elements with color code indicating similar behavior of the metals.

Another difference noted is when Eu and Am are compared. While Eu is a two-electron bonded metal due to its stable  $4f^7 5s^2$  outer configuration), where Am is a three-electron bonded metal ( $5f^6 6d 7s^2$ ). Then a major difference is found for Es – No elements, where these elements are assigned as having two bonding electrons, comparable to the case of Yb, which also has two bonding electrons ( $4f^{14} 5s^2$ ). The Eu and Yb cases are due in part to the stabilization afforded by half- and full filled 4f states. For the Es -No elements it has been shown that their two-electron bonded metallic states result from the greater differences between their f-electron promotion energies for their f-states (e.g.,  $f^n s^2$  to  $f^{n-1} ds^2$  state). The behaviors of these elements in solids and solutions have been determined in other work by many researchers, and they are different than for their metallic states. For example Es and Fm display plus three states in solids and solutions, and Es can also be tetravalent in solids. The case for Lr is special; predictions suggest that it may have a  $p_{1/2}$  outer configuration due to relativistic effects rather than an  $f^{14} ds^2$  state like Lu. Thus, Lr is shown as in the figure as probably having three bonding electrons. Some limited solution studies on a few atoms of Lr have suggested it has a plus three state.

The above information led to our efforts to probe the science of the Es and Fm elements, using electron diffraction and enthalpy of sublimation experiments – both explore the electronic makeup of the metals. Electron microscopy and electron diffraction studies on both Es oxide and metal have been performed, which allowed crystallographic information to be obtained for them via electron diffraction techniques. Conventional X-ray diffraction efforts failed due to the rapid destruction of their crystal matrices by the larger quantities needed. The difference for success with the electron diffraction lay in the very small sizes of the Es particles, which allow the major portion of the alpha, heat and the recoiling nuclei from Es to pass through the very small particles rather than being deposited in their matrices.

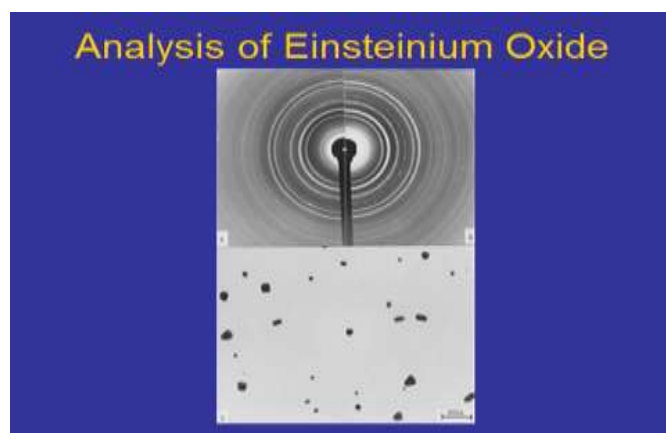


Figure 2. Electronic diffraction pattern from  $Es_2O_3$  nanogram-sized particles.

Electron diffraction (as compared to that of  $Gd_2O_3$ ; left side) and the small nanogram-sized particles of  $Es_2O_3$  from which the diffraction data were obtained are shown in Figure 2.

Similarly, electron diffraction patterns of Es metal were also obtained (not shown here) and they demonstrated that Es is a divalent metal (e.g., having two bonding electrons). Excellent fcc diffraction patterns were obtained for small amounts of Es metal deposited on electron microscopy grids. The diffraction patterns for Es were very similar to those obtained for the divalent, fcc crystal structure



of Yb metal. In further work on Es metal, the preparation of some 120 micrograms of it was performed to study the metal's high-pressure behavior, to determine if pressure would bring about changes in bonding (e.g., force the formation of a trivalent Es metal product by providing the necessary electron promotion energy to form a  $ds^2$  state). See Figure 3 for the sample obtained; the Es is the small piece in the center of the Pt dimple. This probably represents the only known preparation of Es metal, other than the very small quantities prepared for the electron diffraction work mentioned above. The Pt disc was to serve as a pressure cell for the high-pressure study. Unfortunately, the pressure study on this Es metal was unsuccessful, as both the Es and Pt became amorphous rapidly due to the intense radiation and recoiling nuclei from the larger mass of Es.



Figure 3. Image of Es metal.

The enthalpy of vaporization of a metal is a reflection of its electronic bonding. Thus, the diffraction data and the derived metallic radius for Es, plus the enthalpies of vaporization for both Es and Fm metals (see Figure 4.) helped confirm the assignments that these two elements are two-electron bonded metals, just as found with the Eu and Yb metals in the 4f electron series.

The assignment of Md and No as also being two-electron bonded metals (shown above in the two f electron series plot) is based on

calculations and assignments of their f-electron promotion energies (e.g.,  $f^n s^2$  to  $f^{n-1} ds^2$  states) – experiment crystal data and enthalpy of vaporization are not known for them (see discussion below). Nor is the crystal data known for the last metallic element, Lr. Given its short half-life and that only a few atoms can be prepared, experimental information for Lr is very limited, as noted above. A monovalent bonding state has been proposed for Lr based on potential relativistic effects, while some very limited experimental work suggests it could be trivalent ion in solution. Thus, the state of Lr metal presented in Figure 4 is assumed to be a trivalent metal, but arrows are shown for it to move from being “divalent” to have a lower or higher metallic valence.

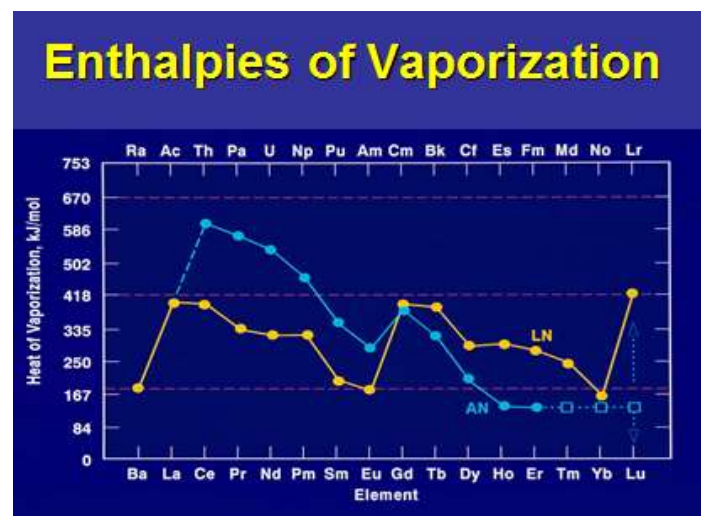


Figure 4. Enthalpies of vaporization of lanthanide and actinide metals.

Other efforts to explore the bonding in Es and Fm metals were attempted by measuring their enthalpy of sublimation via a special technique. These data were done in conjunction with collaborative studies measuring the enthalpies of sublimation/vaporization for the Am, Cm Bk and Cf metals. The enthalpy of sublimation can be used as a measure of the degree of bonding in metals – the greater the enthalpy, the greater the degree of electronic bonding in the metal.

While Cf metal has a depressed enthalpy, it is still larger than compared to those for Es, Fm, Eu and Yb, (see Figure 4). Thus, Cf is believed to normally be a three-electron bonded metal. Thus, the others (Es-No) can be assigned as being two-electron bonded metals. These data help confirm that Es and Fm are indeed “divalent” metals” (e.g., each having two bonding electrons).

Thus, the diffraction data and derived metallic radius for Es, and the enthalpies for both Es and Fm metals all helped confirm the assignments that these two elements are indeed two-electron bonded metals, just as found with the Eu and Yb metals for the 4f electron series. The assignment of Md and No as also being two-electron metals (shown above in the two f electron series plot) is based on calculations and assignments of their promotion energies (e.g.,  $f^n s^2$  to  $f^{n-1} ds^2$  states) for them – experiment crystal data are not known for them, nor for Lr metal. Given its short half-life, only a few atoms of Lr can be prepared, experimental information for it is very limited. As started above, a monovalent bonding state has been proposed for Lr based on potential relativistic effects, but some very limited experimental work suggests it could be trivalent ion in solution.

The above comments on actinide studies and other research studies accomplished in collaborative work with many scientists all reflect a great deal of satisfaction. Hopefully, the results to date have advanced our understanding of the actinide series. Such research should continue and needs to be done. The HFIR is still functional regarding generation of isotopes for research, as well as it being used for different types of neutron studies. This was and remains an important program for the Department of Energy, and for many research institutions. The development of NUCL certainly helped spur

research on these elements, and recognition of its role in the past 50 years in this area of science should be recognized. And the efforts of the Division should be continued.

The Division’s efforts and the results obtained from studies and theory are certainly beneficial in many ways, including the generation of new scientists and attracting students into this arena. And finally, the collaborations and interactions of scientists in such studies cannot be emphasized enough for training, as well establishing important science for understanding and expanding knowledge of the Periodic Table.

With regard to the Periodic Table, there is much more to learn (e.g., “an extended Periodic Table”) – perhaps a new challenge for NUCL). Given some 118 known elements – where IUPAC defines an element for those products with a lifetime of greater than  $10^{-14}$  seconds – one can speculate about the nature of elements above 118. It would seem elements 119 and 120 may have 8s electrons, but then the assignments of the electrons for higher elements becomes a problem. Does one now consider the generation a 6f series or a 5g block of electron grouping for them? And this decision becomes even more difficult when potential relativistic effects are injected!

For me personally, these actinide studies and the many collaborative interactions were not only beneficial but enjoyable, and generated many scientific friends. This 50 Anniversary of NUCL offers a glimpse of the progress and benefits of such an organization, and the 50<sup>th</sup> Anniversary booklet generated will be an important progress report of some of the science accomplished during this period. Congratulations to the NUCL Division on its 50<sup>th</sup> Anniversary.

## So What Do You Get for Being a Member of the Division? 1986-present Michael Bronikowski



*Michael Bronikowski was born in Wurtzberg W. Germany in 1962. He obtained a B.S. degree in chemistry from Marquette University in 1984 and a Ph.D. in Nuclear chemistry at Purdue University under Norbert Porile in 1994. Michael followed this with a post doctoral appointment (1995-1998) working in Gregory Choppin's laboratory at Florida State University. Michael joined the staff of the Savannah River Laboratory in 1998 and presently works at the Savannah River National Laboratory. He has been a member of the division since 1986 and has held the NUCL positions of vice chair (2009), chair elect (2010), chair & program chair (2011) and past chair (2012).*

At the 249th ACS national meeting, Monday night right at the end of the [NUCL] business meeting just before the councilor hands out the drink tickets, always with much aplomb, I was asked this question; "So what do you get for being a member of the Nuclear Division?" I think the question had something to do with the return one gets on the divisional dues but the quick answer was being handed out. "You get a free drink and food at the social hour."

True since I can remember but not a good answer for someone who has been a member for a while. "We used to get the division abstracts mailed to us in the '80's," someone chimed in. "You can get that on line" was the questioner's response. The discussion continued, apparently you can interact and get a lot of things free on line from the Division. The question is not as easy to answer as one would think but I have been in the Division long enough that I wanted to do better than, "You get to communicate".

Norbert Porile, my major professor at Purdue, said it would be useful to become a member of the Division of Nuclear Chemistry and Technology, DNCT, now NUCL, when I joined his group. This was in the '80s when the decline of nuclear chemistry was starting to become apparent. The Nuclear Summer School had recently been started to address the problem. From a young scientist's perspective, the Purdue chemistry department had over 400 grad students but only seven were in the nuclear chemistry program. It was like this around the country. I still remember getting the abstracts in the mail. Here were a bunch of talks I was interested in! Not organic, not p-chem, or analytical, but nuclear reactions and radiochemistry.

As any young member will tell you, they will always remember their first ACS meeting and talk. Mine took place in New York in a long thin room which barely held the audience. There was chatter that Seaborg and even Turkevitch would be there! Jim Cummings was talking after me! Walt Loveland, was presiding over the session that morning. This meant it was his job to make sure a question was asked if the audience didn't. My talk was

on the reaction cross sections of 14.6 GeV/nucleon  $^{16}\text{O}$  on Ag, an ultra-relativistic heavy ion reaction. Walt's question was about whether I would like to do more work on the  $^{102}\text{Rh}$  reaction cross sections. The answer was simple, my thesis is almost done, it would be interesting but  $^{102}\text{mRh}$  and  $^{102}\text{gRh}$  have half-lives of 207 d and 2.9 yr., so, no. The rest of the meeting had discussions of heavy ion reactions, multifragmentation, and the nuclear equation of state, all things one doesn't usually talk or argue about in public. I'm sure some of you don't ever talk about these things because NUCL has another side to it which I found out when I joined Greg Choppin's group. They did radiochemistry. As a former division chair, Greg was very supportive of the Division and had learned to delegate. He wanted you to participate, join a committee or preside over a session, and meet people. This is where you learn presiding over a session means you have to keep the speakers on time, per the ACS, as well as asking a relevant question. It's also where I learned that many of the volunteers that I met who I didn't know were part of the "Choppin Mafia" for having gone through Greg's lab.

I joined Savannah River Laboratory, now SRNL, in the Actinide Technology Section (ATS) to work on separations and process chemistry for the canyons and HB-line. NUCL now seemed to be shrinking as membership was in the mid 800's. The process chemistry I was learning seemed to always lead to overviews in the ACS symposium series from the I&EC or NUCL. Some of the authors like Major Thompson or Dave Karracker still worked down the hall! When we started to work on Np processing, the Savannah River Np processing history from Clark Ice, Don Orth, and others was part of the Division's early history. You can understand why they set the Division up. The work being done didn't really fit in industrial, inorganic, p-chem, or separations chemistry because it was an agglomeration of all of

these. NUCL was now where nuclear and radiochemistry could be discussed.

In 2007, Heino Nitsche, when he was chair, asked me to participate in working on the latest ACS thing, a divisional strategic plan. Heino thought we as a division needed to communicate with everybody in any way possible, especially the way that the younger members do. Remember the Blog? He had amazing enthusiasm and made the discussions flow. He encouraged us to go through the nuclear chemistry vs radiochemical number of symposia discussion, the ever present question of whether the division was DNCT or NUCL, and how we could get more members. He excitedly, put out the plan to the members as an opportunity for them to take part.

It took a few years but I did take part by running for Chair as they needed a second person to run in the election. Don Orth encouraged me to run as he thought that being past chair of the Savannah River local section would be helpful. What he and those who convinced me to run neglected to say was that, it's a ton of work if you win. You get a lot of help from the ACS and the NUCL executive committee, especially the past chair and Frank Kinard. Silvia Jurrison was Past Chair in 2011 and was phenomenally helpful. The radiopharmaceutical symposium she helped set up seemed to reintroduced the discipline to our division and added to our members. Frank was our secretary. He was secretary so long (1996-2013) that no one used his last name. Frank was the guy who lectured in the summer school in San Jose, the one who put the ACS yearly division report together, the one who did business meeting notes, the one who knew when things were supposed to happen, and the one who gave out the coveted Past Chair pin. NUCL honored him recently by establishing the W. Frank Kinard Distinguished Service Award.



In 2011 I became Chair and was asked more than once the question of what do you get? As Chair you get to ask people to help out with things and they generally do it! For example, the Analytical Chemistry in Nuclear Chemistry symposium organized by Coleman, Hobbs, Hobart, Cho, et al., over twelve times since 2000 may be due to the new chair realizing this. My thanks to everyone who helped out especially since 2011, the International Year of Chemistry. As this is recent history I will only note a few things that happened that year. The new PACS system was used to set up symposia, you had to be patient as with a 486 personal computer with the turbo button not on to get it to run. I think this is one of the reasons why the Chair Elect, Ken Nash, made sure the Division get a much needed Program Chair. David Morrissey won the Seaborg Award and Sherry Yennello won the Olin Award, both had great symposia. The symposia "Actinide Materials: Complexity, Nano structural, and Extreme Environments" brought with it many talks and new members. The Fukushima accident happened just before the spring meeting, and the Division had a talk on it. The 100th anniversary of Marie Curies Nobel prize in chemistry was celebrated by a well attended symposia and an

evening rendition of "Marie Curie One Woman Dramatic presentation" with a chocolate desert bar courtesy of the French Consulate.

Two issues came up that year in the Business Meeting, both of them still affect NUCL. The first is that NUCL had 993 members and I wanted 1000. I wish I could say that I gave an impassioned speech to add 7 more members, but I think Darleane Hoffman's, "We can easily do that" remark helped more. With over a thousand members, NUCL now has a second councilor. The second is that we had to address the Seaborg Award. The Division needed to work on strengthening nominations and the funding. I think now only the funding issue still needs some work as part of your dues pays for it. This brings me back to the question at hand.

What do you get for your membership? Is it useful information, participation, meeting people, talk about things you don't talk about in public, or pay to honor great chemists? Maybe you get to meet great chemist like Al Ghiorso. My answer is; you get to interactively communicate with a group of people who have the same interests to make some great memories. Happy 50<sup>th</sup>, NUCL!

## Evolving Professional Preferences

Corinne Dorais



*Corinne Dorais is a second-year Ph.D. student at the University of Notre Dame. She is co-advised by Professors Amy Hixon and Antonio Simonetti. Her research focuses on method and standard development for both pre- and post-detonation nuclear forensics applications.*

Growing up, I knew I was going to be an astronaut. I was so certain of this fact, that on our sixth birthday I elicited a very solemn promise from my brother that we could be the first twin astronauts in NASA. Though we later decided it more prudent that he remain on Earth due to a rather common six-year-old affliction (a fear of heights) my dreams remained in the stars. While I admit that the idea of being able to jump higher than I ever could on Earth was a primary motivator, I was fascinated by the idea of discovering something new, of figuring out how our universe worked. My parents encouraged this exploration, building baking soda and vinegar volcanoes with me in the backyard and watching the crystals of rock candy creep up a string.

As I moved into junior high school, my career goals began to shift. A newfound love of biology and a burgeoning desire to use my career to help others steered me towards medicine. This new dream lasted until early in college when I discovered a love for physics.

The more time I spent in school, the more I realized I would be happiest in a research career. The question then became, which field? Physics? Biology? Environmental Sciences? Anything seemed possible. Anything, that is, except for chemistry. Chemistry had never captured my attention the way other sciences did. Sure, it was interesting, but there was so much memorization involved that I never saw beyond it to synthesis, research, or exploration of new material.

After graduating from Juniata College with a B.S. degree in Environmental Sciences, I spent a year working in Professor Patricia Holden's environmental microbiology laboratory at the University of California, Santa Barbara. Under Dr. Holden's guidance and mentorship, I decided to apply to Ph.D. programs in Environmental Engineering. So it was that I applied to several universities, including the University of Notre Dame. A short while after submitting my application, I received an email from Dr. Amy Hixon, an assistant professor in the Department of Civil and Environmental Engineering and Earth Sciences. She had seen my application and wanted to know if I had any interest in radiochemistry, a field I promptly had to Google. Radiochemistry, I worried, sounded an awful lot like my least favorite science. And yet as Dr. Hixon explained more on the phone and later in person, it sounded fascinating. Suddenly my easy graduate school decision did not seem so easy any more as I was torn between this new field of radiochemistry or one of the other programs in environmental engineering. The decision was a long one, full of pros and cons lists and my patient parents listening to me agonize over every detail. Ultimately, I chose to work with Dr. Hixon at the University of Notre Dame; I am so glad I did.

I started my Ph.D. program in the fall of 2014, taking classes such as Actinide Chemistry while beginning my research. My preliminary work focused on uranyl peroxide cage cluster interactions at mineral interfaces. These cage clusters have been shown to self-assemble in aqueous solution under conditions similar to those expected to exist around high-level radioactive waste, such as spent nuclear fuel. Given the potential for these clusters to form in contaminated environments, it is necessary to understand the radiochemical and geochemical factors that influence their behavior. This work will be presented in December 2015 at the Pacificchem conference in Honolulu.

My research has since shifted to method and standard development for both pre- and post-detonation nuclear forensics applications. With Dr. Hixon, I am working to create and characterize a surrogate material for post-detonation nuclear debris that can be used for the quantitative analysis of actinides and fission fragments in the event of an urban nuclear detonation. In February 2015, I began to be co-advised by Professor Antonio Simonetti. My work with Dr. Simonetti is focused on the development of natural standards for nuclear forensic analysis that can be used to determine and validate chemical and isotopic signatures at high spatial resolution.

In order to learn the chemical and analytical techniques necessary for this research, I spent summer 2015 as a Seaborg Fellow working with Dr. William Kinman at Los Alamos National Laboratory (LANL). There I learned anion exchange column chromatography for trace-level uranium and plutonium separations. My time at LANL convinced me to pursue a career at a Department of Energy laboratory and I hope to return to work at LANL to gain more experience prior to completing my dissertation.

From LANL, I traveled to the International Atomic Energy Agency (IAEA) in Vienna, Austria to attend the annual meeting of Women in Nuclear Global. This meeting, attended by women from around the world in a variety of fields relating to the nuclear sector, reinforced in me the need for international collaboration and cooperation in nuclear energy, security, and safety. In addition to fostering international dialogue and networking, the meeting facilitated tours of relevant nuclear-related sites such as the IAEA's Incident and Emergency Center, which coordinates the international response to nuclear and radiological crises; the Zwentendorf nuclear power plant, which never went critical and therefore allows visitors the opportunity to stand inside the reactor core; and the Atominstytut, Austria's only operational nuclear reactor.



Figure 1. Author at Zwentendorf, the Austrian nuclear power plant that never went critical.

Since my return to Notre Dame at the beginning of the fall 2015 semester, I have been working to establish the chemical separations and subsequent analyses I learned at LANL. I recently joined the

editorial board for Frontiers in Energy Research, a newsletter showcasing the research conducted within the Department of Energy's Energy Frontier Research Centers. Additionally, I am working to finish the last of my coursework and prepare for my qualifying examinations, which will be held in January 2016.

Five years ago, I would have never predicted I would be pursuing a PhD in radiochemistry, yet it has been an incredibly rewarding year and a half. I am eager to see where the next

five years in this field lead. Happy 50th Anniversary NUCL!



Figure 2. Inside the reactor core at Zwentendorf



# History of the Division of Nuclear Chemistry and Technology's Summer Schools in Nuclear Chemistry

Trish Baisden



*Trish Baisden earned her Ph.D. in Chemistry in 1975 from Florida State University under the direction of Prof. Gregory R. Choppin. After receiving her Ph.D., she spent two years as a postdoctoral fellow with Prof. Glenn T. Seaborg at the University of California, Lawrence Berkeley Laboratory studying the chemical and nuclear properties of the heavy and super heavy elements. She then joined Dr. Ken Hulet's Group as a staff member at the Lawrence Livermore National Laboratory (LLNL) where she investigated fission properties of the actinides. During her over 35 year career at LLNL, Trish held a number of technical management positions including Division Leader, Deputy Director of the Seaborg Institute, and Deputy Associate Director for Chemistry and Materials Science. Toward the end of her career she worked on developing large optics for the National Ignition Facility (NIF) and when the laser was completed, she served as the Deputy Director of the National Ignition Campaign, a multi-laboratory scientific effort on the NIF to use inertial confinement fusion to achieve ignition and thermonuclear burn in the laboratory. Trish retired from LLNL in 2013 and became the primary instructor for the DOE/ACS*

*Summer School in Nuclear Chemistry at SJSU.*

In 1977, the ACS Division of Nuclear Chemistry and Technology (DNCT), now NUCL, Chairman, Bruce Dropesky (LANL) formed an ad hoc committee to conduct a national survey on the status of training of nuclear and radiochemists. The committee included Rolfe Herber (Rutgers), Glenn Gordon (U. of Maryland), Dave O'Kelly (ORNL) and was chaired by Greg Choppin (Florida State University). The key result from the survey<sup>1</sup> was that a significant increase in demand for nuclear and radiochemists was expected in the 1980s but a declining student population would not provide adequate graduates to meet the need. Evidence cited for this claim was the nearly 60% decrease in the number of Ph.D. degrees awarded in 1976 as compared to that projected in 1980-81. The committee also noted the



Figure 1. Greg Choppin (left), Trish Baisden (center) and Gerhart Friedlander (right) circa 1980 discussing the idea of a summer school at an ACS meeting.

<sup>1</sup> ACS DNCT (now NUCL) , Report of the ad hoc Committee on Training of Nuclear and Radiochemists, 1978  
NUCL Newsletter, 50<sup>th</sup> Anniversary, Page - 29

problem would be exacerbated due to expected retirement of a number of nuclear and radiochemistry faculty at US colleges over the same period of time. The committee concluded that a sustained effort would be necessary to stimulate growth of student interest and reverse the trend of declining faculty. Further, the committee stated that “such stimulation could be achieved only by the direct action of federal agencies through fellowships, increased research funding and the addition of younger faculty in nuclear and radiochemistry.” This issue created a lot of talk within NUCL and it was generally agreed that funding at the time was marginal for the current level of faculty and students and certainly inadequate for any growth. Filling the pipeline was also seen as a big problem because each year it was noted that the number of undergraduate chemistry programs offering a course in nuclear and radiochemistry was shrinking rapidly. Having received my Ph.D. from Prof. Choppin, I had the opportunity to discuss the problem at length with him. Greg was very passionate about teaching and ensuring the health of the field and he inspired me to become part of the solution. He told me to get involved and serve on the NUCL Committee on Training.

At the time, I was a postdoctoral student with Glenn Seaborg at UC Berkley and looking for a way to become involved in the NUCL Division. I discussed this with Prof. Seaborg and he likewise encouraged me to step up.

While at Berkeley, I saw a poster for a topical summer school in Europe and it started me thinking....a great way to stimulate student interest would be through a Summer School in Nuclear and Radiochemistry. The summer school would be an intensive, 6-week lecture and laboratory course designed to introduce outstanding upper level undergraduate science and engineering majors, selected through a national competition, to nuclear and radiochemical concepts. In addition, we

wanted them to meet and be inspired by people working in the field through a Guest Lecture Series and visit nuclear science facilities where nuclear research was being conducted before they made their minds up about what to pursue in graduate school. Students completing the program that showed interest in the field would be helped to find summer employment the following year and be given help with finding and applying to graduate program in nuclear and radiochemistry. At the time there weren't many summer programs for undergraduates and none of the existing programs targeted US citizens and offered the cradle-to-grave attention to the students we were proposing. The more I thought about it, a Summer School in Nuclear and Radiochemistry, a fellowship program for undergraduates, the better it sounded. However, there were a lot of details that needed to be worked out and a lot of people that needed to be convinced. The first hurdle was to answer the question “was this an appropriate activity for NUCL, and if so, as an all-volunteer organization could we pull it off?” After all, we were just a professional society.

About this time, I completed my postdoctoral appointment and moved to LLNL as a staff scientist working in Ken Hulet's Heavy Element Group within the Nuclear Chemistry Division (NCD). One of the first thing I did at LLNL was to engage Christopher Gatrousis, the NCD Division Leader with the idea. He encouraged me to pursue the summer school idea and offered to help. So I prepared a proposal that described in detail the concept and the elements needed to make it successful including a process for evaluating, assessing and providing feedback to the program to improve content and quality of the summer school.

From 1981 through early 1983, I pitched the idea to anyone who would listen, not the least of which were the NUCL Chairs and

Executive Committees. I also approached the then Standing Committee on Nuclear and Radiochemistry (CNR) of the National Academy of Sciences/National Research Council (NAS/NRC). Ed Macias (U. of Washington, St. Louis) was the committee chair and Bill Spindel was its NRC Staff Officer. Ed Macias, as well as other members of the committee, in particular Gerhart Friedlander and John Huizenga, were somewhat skeptical that an all-volunteer organization such as NUCL could be an appropriate sponsor for an educational program. Darleane Hoffman, also a member of the committee, was very encouraging and told me in her usual way not to stop until I had “worn the guys down and they agreed.” One of the concerns of the committee was that if the summer school was held at a university with a Ph.D. program, it would become a feeder program for that school. Things changed when they learned that the plan was to hold the summer school at the San Jose State University (SJSU) because they had: 1) appropriate facilities and instructors to carry out the program (Profs. Alan Ling and Ruth Yaffe and the SJSU Nuclear Science Teaching Facility), 2) no graduate program in chemistry, 3) the ability to offer college credit and affordable student housing (International House), and 4) was conveniently located in the San Francisco Bay Area with easy access to several national labs with associated “nuclear science facilities” (e.g, LBNL, LLNL, Stanford, NASA). By mid-1983, with my promise to personally handle all of the administrative issues and the associated paperwork, everyone was on board including the NUCL Executive Committees and Chairs (1981) Jerry Hudis, BNL, (1982) Dick Hahn, ORNL, and (1983) Shelly Kaufman, ANL, and the NAS/NRC CNR. We even got Barbara Hodsdon and Christine Pruitt in the ACS Division Activities Office to agree that because this was an educational program, if we were successful obtaining funding, we could channel the grant funds through the ACS with

no overhead as long as we prepared the required progress and financial reports.

The next hurdle was to convince a federal agency to fund our Summer School. The obvious place to start was the Department of Energy (DOE) since they are responsible for the effective stewardship of 17 world-class national laboratories and they acknowledge “Applied Nuclear Science and Technology” as a core competency necessary to execute their missions. Enter, Elliot Pierce, a long standing member and councilor of NUCL and Director of ER-14, in the DOE Office of Basic Energy Sciences. Elliot took the proposal to his boss, Antoinette Joseph, along with a letter excerpts of which are below:

“This proposal is so impressive, compared to the many educational proposals that cross my desk that I am compelled to urge your full consideration of it. This is the only time I have made a recommendation of this sort.”.....“This proposal is a stand-out, it has all the virtues one could hope for, some of them unique: serving a set of serious national needs, a plan carefully thought through, a highly competent and enthusiastic staff, an excellent and unique facility, unique access to the best scientific and educational advice, careful selection of students and unusual attention to later building on the student’s summer experience.”

The next thing we knew, we were funded for 1 year with the possibility for a second for a total of \$50K plus a \$5K grant from Exxon Foundation.



Figure 2. Photo of Elliot Pierce. “Without Elliot Pierce’s help, attention, and persistence, an educational outreach program such as embodied by the summer school would never have gotten off the ground” – Trish Baisden.

The next step was to solicit student participants. Bill Spindel offered to run the application process through his office and it was his assistant Peggy Posey that handled the administrative paperwork for the first year. Announcement fliers (1400) were designed, printed, and mailed gratis by LLNL to all US colleges and universities having a chemistry or physical sciences department and to all ACS Students Affiliate's Chapter (ACS mailing list). A selection committee composed of Ed Macias (CNR) and Rolfe Herber and Dick Hahn (NUCL) waded through the 71 competed application packets and selected the top 12 students and alternates. It was decided to make offers to 14 students and to our surprise, all accepted. The first class consisted of 11 students who had completed their junior year and 3 sophomores, 11 males and 3 females. The students selected for the first class, the "Class of 1984" were:

Carolyn Anderson –U. of Wisconsin-Milwaukee

Carlos Barbas – Eckard College

Kristie Boring – U. of California – San Diego

Terry Coley – U. of Illinois-Campaign-Urbana

Dan Dorsett – U. of California – Berkeley

John Gingrich – U. of Indiana – Purdue

Howard Hall – College of Charleston (SC)

Claire Huiras – U of Miami (Florida)

Dan Jones – U of Pittsburg

Howard Lee – Harvard University

Michael Mohar – U of Chicago

Karl Mueller – U of Rochester

John Circle – Butler University

Mark Stoyer – Purdue University

As advertised, the first summer school was an intensive program. A typical day at the summer school started at 8 am with lectures until noon with afternoon labs from 1- 5 pm.

Evenings and part of the weekends were spent working problem sets, writing labs reports, studying for quizzes and exams, and preparing a presentation on a self-selected nuclear topic. Saturdays were reserved for rest and relaxation and excursions such as a

San Francisco bay cruise, visits to a redwood forest and the beaches at Santa Cruz, or the aquarium at Monterey Bay.



Figure 3. Carlos Barbas and Dan Jones (Class of '84) working on a lab experiment in the counting room at SJSU.

The students worked hard and they learned a lot. They enjoyed time out of the classroom on field trips to local laboratories, and they especially relished meeting and interacting with the Guest Lecturers. The Guest Lectures for the first summer school were Glenn Seaborg—"New Alchemy", Ed Macias—"Radiochemical Techniques and the Environment", Vince Quinn—"Neutron Activation and Crime Investigation", Greg Choppin—"Nuclear Power, Fuel Reprocessing, and Waste Disposal", and Hal O'Brien—"Nuclear Medicine."



Figure 4. Summer school students relaxing and taking in a breath taking view of the SF Bay on a Saturday.





Figure 5. Prof. Alan Ling getting to know the 1984 class at the International House.

By the end, everyone was exhausted, students and instructors alike, but we had accomplished what we set out to do. In jest, the students fondly referred to the program as the “Nuclear Boot Camp” and the name stuck. Howard Hall was selected as the “Outstanding Student” and was given a trip to the national ACS meeting held that fall in Philadelphia. At the NUCL business meeting, Howard described his experience at the Summer School and was given an award. An amazing thing was that five of Howard’s summer school classmates commandeered a van and drove across country to see Howard get his award.



Figure 6. Prof. Gregory Choppin lecturing to the first summer school at SJSU in 1984.

The year following the first summer school students were provided assistance with graduate school applications which included writing reference letters. For those expressing interest in the field, we helped them either find graduate program matching their interests or summer employment doing research in nuclear or radiochemistry. This was a particularly important aspect of the summer school which we have continued to

this day. During the same time, I provided DOE with the progress reports they needed while Joe Peterson, the NUCL Treasurer, handled the associated financial reporting. DOE seemed very happy with the results and requested NUCL write a multi-year proposal for funding. We did so and we were off and running.



Figure 7. Howard Hall, the Outstanding Student of the 1984 class attending the Philadelphia ACS meeting in the fall of 1984.

From the second year on, the application and selection processes were carried out by NUCL and later a two day module “Workshop on Nuclear Medicine” was added. Organizers of the Nuclear Medicine Workshops over the years at SJSU included Hal O’Brien (LANL), Mike Welch and Carolyn Anderson (Washington University, St. Louis) and Henry Vanbrocklin (UC San Francisco). These workshops also included tours of facilities used to produce radioisotopes and synthesize radio-pharmaceuticals for imaging and therapy at UC San Francisco



Figure 8. Henry Vanbrocklin (center) describing to the 2012 class the use of the cyclotron at the UCSF nuclear medicine facilities to produce radioisotopes for PET imaging.



(Henry Vanbrocklin) and Stanford (Fred Chin). In 1987, NUCL won its first ACS Award for Outstanding Small Division primarily for the establishment and successful operation of the summer school as a significant educational and outreach project. In 1988, the NAS/NRC held a workshop<sup>2</sup> again addressing the topic of the training of nuclear and radiochemists. A key recommendation from the workshop was to establish a second summer school in nuclear and radiochemistry at an eastern site. Fortunately, about a year earlier, I was approach by Dr. Seymour Katcoff about BNL being the site for a potential second summer school. I thought it was a good idea and encouraged him to work out the details with the BNL management. Then when the workshop report came out, BNL was well on its way allowing me to approach DOE about funding. They were fully supportive so the next cycle of the program grant was written to include a second summer school.



Figure 9. Seymour Katcoff circa 1988 was responsible for establishing the second summer school at BNL.

The first summer school at BNL was held concurrently with the SJSU program in the summer of 1989. Seymour Katcoff was the BNL Site Director and the main instructors for the BNL site were John Alexander (SUNY, Stony Brook) and Daeg Brenner (Clark U.). The laboratory portion of the course was set up and taught by Ivor Preiss (RPI) and RPI is credited with loaning some equipment to BNL to accommodate a student laboratory. Other

key personnel from BNL included Y.Y. Chu who helped with the laboratory. Later in the program Y.Y. Chu also was the BNL Site Director and primary lecturer along with Peter Haustein (BNL) and Roy Lacey (SUNY Stony Brook). Analogous to the SJSU program, BNL had a very successful Guest Lecture Series, Nuclear Medicine symposium (organizers at BNL included Joanna Fowler, Len Mausner, and Kathryn Kolsky) and tours of local nuclear facilities including an operating nuclear power plant.



Figure 10. BNL 2011 Class visiting the control room of a nuclear power plant.

It is now the fall of 2015 and the Summer Schools have been in existence for 32 years and included 699 student participants (since 1984 at SJSU- 383 and since 1989 at BNL – 316). With this longevity, it begs to ask the question, “Why have the Summer Schools worked?” The answer is multidimensional. The program has attracted high quality and very enthusiastic students who are genuinely interested in learning about nuclear and radiochemistry. The students have come from small schools and large schools most of which do not offer undergraduate courses in nuclear and radiochemistry. The longevity of the program has ensured the program is well known across the country. It is gratifying that

<sup>2</sup> Training Requirements for Chemists in Nuclear Medicine, Industry, and Related Areas – Report of a Workshop, Co-Chaired by Gregory. R. Choppin and Michael J. Welch, National Academy Press, Washington, DC, 1988

graduates of the summer schools continue to be our most effective program advocates with other students and with the their home institutions.

Support from the ACS especially in the early years and the continuing moral and financial support from the DOE over the history of the program. We especially want to thank OBES and NUCL Councilor, Elliot Pierce, Office of University and Industry Programs Director, Richard Stephens, OBES Federal Program Managers, Bob Marianelli, Lester Morse, and Philip Wilk (Summer School Class of 1994), and Office of Nuclear Physics (Isotope R&D) Federal Program Manager, Dennis Phillips.

**NUCL members who have participated in the Guest Lecture Series at both SJSU and BNL.** These men and women who have given of their time to provide fascinating seminars and inspire student through personal interactions. There are too many to name but you know who you are.

**NUCL members who have arranged the many tours of the nuclear science facilities at the national laboratories.** Special thanks for SJSU program go to Glenn Seaborg, Darleane Hoffman, Heino Nitsche and David Shuh for organizing the visits to LBNL.



Figure 11. Glenn Seaborg, Darleane Hoffman, and David Shuh (left to right) arranged visits for the summer school to LBNL over the years.

Likewise for SJSU, Annie Kersting and the Seaborg Institute for the visits to LLNL. Special thanks for the BNL program go to Leonard Mausner and Suzanne Smith at BNL, Roy Lacey at Stony Brook University,

and Jason Lewis at Memorial Sloan Ketterling Cancer Center.

**NUCL members who have served as the National Program Directors for the Summer Schools.** They include Trish Baisden (1984-1994), Joe Peterson, U. of Tennessee (1995-2001), Sue Clark, Washington State University (2002-2006), Paul Mantica, Michigan State University (2007-2011), and David Robertson, U. of Missouri, Columbia (2012 to present).



Figure 12. National Summer School Directors (from left) Joe Peterson, Sue Clark, and Paul Mantica.

Although each of these national directors did a tremendous job, the special efforts of Sue Clark to keep the program competitive with summer research position by providing student stipends, and David Robertson to push back on the Obama administration's efforts through the Office and Science and Technology to terminate program funding and reorganize and redirect federal agency-related Science, Technology, Engineering and Mathematics programs.



Figure 13. David Robertson (right), current National Program Director on a site with the 2013 class at SJSU.

**The Instructors and Site Directors at SJSU and BNL over the history of the program.** SJSU – Alan Ling (Site Director and Principal Instructor, 1984-1993), Peter Englert (Site Director and Principal Instructor, 1994-1995), Bob Silva (Principal Instructor, 1996-1998), W. Frank Kinard (Principal Instructor, 1999-2012), Trish Baisden (Principal Instructor, 2013-present, with help from Dale Ensor and Paul Karol), Herb Silber (Site Director, 1996-2015).



Figure 14. SJSU– Principal Instructors- Bob Silva (left) (1996-98). Frank Kinard (center) (1999-2012), and Herb Silber - Site Director (1996-2015).

BNL - Seymour Katcoff, Y.Y. Chu, Peter Haustein, John Boger (Site Director, 1989-2000), Kathryn Kolsky (Site Director 2001-2006), Rich Ferrieri (Site Director, 2007-2009), Louis Pena (Site Director, 2010-present) and the lecturers Cody Folden, Dave Robertson, Alice Mignerey, Romauldo de Souza, Paul Mantica and Ken Czerwinski. As summer school instructors retired within the BNL program, they were replaced by a variety of different lecturers, each lecturing for one week. For more information on the history of and contributors to both the SJSU and BNL program see the articles written by A. Ling<sup>3</sup>,

J. Peterson<sup>4</sup>, S. Clark<sup>5</sup>, K. Kolsky<sup>6</sup>, and W.F. Kinard and H. Silber<sup>7</sup>.



Figure 15. Heino Nitsche showing Frank Kinard the Heavy Elements Research Labs on the visit of the 2012 SJSU class to LBNL.

Last but by no means least, thanks goes to the student participants giving back to the program by serving as teaching assistants, going on to graduate school pursuing nuclear and radiochemistry as their professional careers, and those that have come back to participate in the Guest Lectures and inspire other students to following in their paths.

Apparently, we did something right and have been cited in both the 2007 Nuclear Science Advisory Committee (NSAC) Workshop Report and in the 2012 NRC Workshop Report “Assuring a Future US-Based Nuclear and Radiochemistry Expertise” as “current best practices.” Since it is said, “imitation is the sincerest form of flattery,” we, NUCL, are flattered by the fact that our Summer School program has been duplicated by the Dept. of Homeland Security in the US and ACTINET in Europe. As to the recent impact of the program, three participants in the 2005 Summer School were recently hired as

<sup>3</sup> A.C. Ling, P.A.J. Englert, and C.A. Stone, Nuclear Science Programs at San Jose State University, J. Radioanal. Nuc. Chem, 171, 167 (1993).

<sup>4</sup> J.R. Peterson, The American Chemical Society’s Division of Nuclear Chemistry and Technology’s summer school in nuclear and radiochemistry, J. Radioanal. Nuc. Chem, 219, 231-236 (1997).

<sup>5</sup> S. B. Clark, The American Chemical Society’s Summer School in Nuclear and Radiochemistry, J. Radioanal. Nuc. Chem, 263, 107-110 (2005).

<sup>6</sup> K.L. Kolsky, Summer School in Nuclear and Radiochemistry at Brookhaven National Laboratory, J. Radioanal. Nuc. Chem, 263, 145-149 (2005).

<sup>7</sup> The Department of Energy/American Chemical Society Summer School in Nuclear and Radiochemistry at San Jose State University, J. Radioanal. Nuc. Chem, 263, 155-158 (2005).

assistant professors in nuclear and radiochemistry programs at the University of Notre Dame, the Colorado School of Mines, and Michigan State University. Twelve of the 24 participants in the 2012 Nuclear Chemistry Summer Schools (50%) are currently pursuing graduate degrees in Nuclear Chemistry, Radiochemistry and Nuclear Engineering. Likewise, eleven of the 24 participants (46%) in the 2013 Nuclear Chemistry Summer Schools class are

pursuing graduate degrees in nuclear science programs.

In closing, I would like to again thank the members of NUCL for their continued support and participation in the Summer Schools Program. Also many kudos to LLNL for their willingness to assist in defraying costs, especially in the early years by designing and printing the announcement posters, providing travel funding to help support the Guest Lecture series, and allowing release time for employees to participate.



## Reflections on 50 Years in Nuclear Science

### Major Thompson



*Major Thompson was born in rural north Alabama and grew up in Birmingham. He obtained his B.S. degree in Chemistry from Birmingham Southern College in 1959. He obtained his M.S. and Ph.D. degrees in Inorganic Chemistry from the Ohio State University in 1961 and 1963, respectively, under the direction of Darryl Busch. While at Ohio State he met and married his wife of 52 years, Mary Ann Roof. He was hired by E.I. DuPont in 1963 to work at the Savannah River Laboratory (now Savannah River National Laboratory). Major retired at the end of 2002 and shortly thereafter returned on a part time basis. Major is an Emeritus member of the American Chemical Society. He received the Westinghouse Savannah River Company Don Orth Award for technical excellence in 1996, the Glenn T. Seaborg Award in Actinide Separations in 1997, and the Distinguished Scientist Award from Citizens for Nuclear Technology Awareness in 2003.*

I arrived at the Savannah River Laboratory in 1963 fresh out of graduate school with no previous experience working with radioactive materials. I learned by working with experienced co-workers and following the procedures for working in glove boxes and radio-hoods. I also had no background in actinide chemistry but tried to get up to speed by reading the literature and consulting

frequently with my colleagues in the Actinide Chemistry Division at SRL. My first research assignment was to develop a method to directly reduce plutonium oxide to the metal using calcium metal in an inductively heated calcium fluoride flux to dissolve the calcium oxide formed during the reduction. The method reduced some but not all of the plutonium oxide. My technical report on this work suggested that addition of calcium chloride to the calcium fluoride flux would improve the yield of plutonium metal. Los Alamos would later successfully deploy direct plutonium oxide reduction by using calcium chloride.

I continued to broaden my work experiences in nuclear and radiochemistry through various assignments at Savannah River. This included nuclear reactor irradiation work and learning how to determine nuclear cross sections from irradiation data. During my 39 plus-year career I learned the chemistry and performed experimental programs on all the actinides from thorium through einsteinium. I was able to support the solvent extraction separations for plutonium, uranium, americium, and curium. I was involved with studies of most the nuclear fuel cycle including high-level nuclear waste chemistry as well as reactors and separations and supported both Savannah River and Hanford with these studies.

Prior to formation of the ACS Division of Nuclear Chemistry and Technology in the mid-1960's, most of my scientific information came from the literature and my SRL colleagues. I joined NUCL in about 1970 and found that this immediately widened my technical resources and contacts. Attending NUCL meetings allowed me to present my own work but more importantly to interact with colleagues throughout the world with



similar interests in nuclear science and technology. I found it a bit of challenge not having any academic background in nuclear science, but being a NUCL member and attending the meetings helped me increase the knowledge base that I needed to perform research in nuclear science.

I would like to join all the others in congratulating NUCL on celebrating 50 years

of service and for the opportunities it has provided for many chemists, physicists, and engineers to interact and share their work with others in the field. I also want to take this opportunity to tell young scientists and engineers that I have and continue to enjoy the work, challenges, and interactions with colleagues over my total of 52 years in nuclear chemistry and technology.

## A Career from Petrology to Nuclear Science

Annie B. Kersting



*Annie was born in Berkeley, CA where she lived until she was ten. Between the ages of 10 and 17 she lived in Iran and Indonesia, where her parents taught at American and international schools. She spent her senior year of high school in San Diego before moving back to Berkeley to go to college. She obtained her B.S degree in geology and geophysics at U.C. Berkeley and her M.S. and Ph.D. from the University of Michigan, Ann Arbor. In 1992, she started at Lawrence Livermore National Laboratory (LLNL) as a postdoc in the Institute of Geophysics and Planetary Physics and later signed on as a staff scientist. She is currently the Director of the Glenn T. Seaborg Institute in the Physical Life Sciences Directorate, LLNL where the focus is to foster collaborative research with the academic community in the areas of radiochemistry, and nuclear forensics. The Seaborg Institute serves as a national center for the education and training of undergraduate and graduate students, postdocs and faculty in actinide science.*

I went to U.C. Berkeley as an undergraduate during the late 70's early 80's and it was an incredibly exciting time to be in college. The campus was dynamic and many students were

politically aware. It was a great time and place to attend college as I was surrounded by very smart, creative, and socially aware people. I always liked math and science but I used to fill out my electives by going to the university book store and reading the required book lists for all different classes and then signing up for what looked most interesting. I took a general geology class and was hooked. Although I didn't double major, I would advise those starting college now to think about getting a broad undergraduate education as nowadays, a lot of science is done at the intersections of different fields.

After graduation I worked as a field geologist for about a year, doing work in Mexico, and Northern California, working for U.C. Berkeley, Union Geothermal, and Chevron while deciding what to study in graduate school. I entered the Ph.D. program at University of Michigan to work with Dr. Richard Arculus. My Ph.D focused on experimental petrology of volcanic rocks with the goal of trying to better understand the chemical evolution of the earth. Graduate school really taught me how to think scientifically, to pose a question, design an experiment and to learn that it was OK to have experiments fail. After my Ph.D., I applied for postdocs all over the U.S., but knew I wanted to go back to California. LLNL offered me a postdoctoral opportunity in the Institute of Geophysics and Planetary Physics directed by Dr. Claire Max. I worked with Drs. George Zandt and Rick Ryerson and spent the next three years examining the isotopic signatures of island arc volcanoes to understand the chemical recycling at convergent plate margins and crustal growth evolution. My work showed that we could use Pb isotopic signatures from island arc volcanic rocks to detect chemical contributions from the lithospheric lower crust even if those

contributions were not detectable in the major and trace elemental signatures of primitive basalts. Although at the time I thought I would go on to an academic job, I was offered a staff position and decided to accept.



Figure 1. Top: Aerial view of the NNSS showing several subsidence craters. (<http://nuclearweaponarchive.org>) Bottom: Drill-back rig set-up to drill into an underground nuclear test at the NNSS, NV for collecting soil and groundwater samples. From left to right: Annie Kersting, Jacqueline Kenneally, Karen Patten and Allen Friensehner.

As a staff scientist, I was part of a team working at the Nevada Nuclear Science Site (NNSS) (formerly the Nevada Test Site) conducting both field and laboratory experiments to understand the behavior of radionuclides deposited in the subsurface after an underground nuclear test. We can use

the NNSS to understand how radionuclides will behave in the environment to both help clean-up other contaminated sites and design a safe geologic barrier for long-term nuclear waste storage. The field and laboratory work involved collecting groundwater and soil samples near underground nuclear tests and determining what radionuclides were present and at what concentrations. On one project, we worked with Los Alamos National Laboratory to measure the unique isotopic signatures of low-levels of plutonium detected in groundwater samples to show that the plutonium had been transported several kilometers. This work was one of the first studies to show that insoluble radionuclides, like plutonium, could migrate as suspended, nanometer-sized colloidal particles. This work changed the way we thought about migration of insoluble actinides. Recent research at NNSS focuses on experiments designed to better understand the chemical conditions controlling actinide migration and modeling the transport behavior. During this time at LLNL, I learned that I liked working in groups, where scientists bring a range of expertise to address difficult problems.



Figure 2. Photograph from the 2014 Nuclear Forensics & Environmental Radiochemistry summer program.

I am currently the Director of the Glenn T. Seaborg Institute where we conduct collaborative research with the academic community in the areas of radiochemistry and nuclear forensics. In addition, we help educate the next generation of nuclear scientists

through our graduate and undergraduate student summer programs. Our summer program, funded by the Department of Homeland Security, focuses on nuclear forensics and gives graduates and undergraduates an opportunity to come to LLNL for a hands-on research experience to conduct research with staff scientists. The goal of the nuclear forensics summer program is to train the next generation of nuclear scientists and engineers to solve critical national security problems and have the students participate in conducting research at LLNL. This program has been a successful pipeline for future staff scientists by bringing in graduate students and then later hiring them as postdocs and eventually staff scientists.



Figure 3. Photograph from the 2015 Environmental Radiochemistry Research Group consisting of staff, postdocs and visiting graduate students. Left to right: Annie Kersting, Jennifer Wong, Mark Boggs, Adele Panasci, Harris Mason, James Begg, Chad Durrant, Claudia Joseph, Mavrik Zavarin, Brenda Rubenstein, Tashi Parsons-Moss, Pihong Zhao, Ruth Kips and Yongqin Jiao.

Most of my current research within the Seaborg Institute is focused on identifying the dominant bio-geo-chemical processes and the underlying mechanisms that control actinide transport (focusing on Pu and Np). For plutonium, we are determining what processes occur at both low-level environmental concentrations and at the nanoscale--at the mineral-organic-microbial-water interface that ultimately controls the behavior in the subsurface. Our goal is to be able to reliably predict and control the cycling and mobility of actinides in the subsurface.

Looking back over my career in nuclear science I didn't follow a straight career path. I always took advantage of jobs, projects, and opportunities that interested me. I have been offered amazing opportunities to work on interesting problems with talented scientists. I certainly like the work that I have done in nuclear chemistry and really wouldn't have it any other way.

The NUCL Division of ACS has an outstanding record of providing opportunities for education, networking, and outreach. It is a great time to be involved in nuclear chemistry and the NUCL Division as we have many interesting, scientifically challenging, problems that need solutions from the next generation of scientists.



# From Nuclear Chemistry to Nuclear Medicine: The Role of NUCL Chemists in Medical Imaging

Suzanne E. Lapi and Alan B. Packard



*Suzanne Lapi graduated with her PhD in nuclear chemistry from Simon Fraser University. Following this she worked on development of new molecular imaging agents for prostate cancer at UCSF. In 2008 she joined the faculty at Washington University in St. Louis as director of isotope production where she worked closely with Dr. Michael Welch until his passing in 2012. In 2015 she moved to the University of Alabama at Birmingham as Cyclotron Facility Director and Associate Professor in Radiology. Suzanne's research aims to produce and characterize novel positron emitting isotopes for molecular imaging including  $^{64}\text{Cu}$ ,  $^{89}\text{Zr}$ ,  $^{52}\text{Mn}$  and others.*

*Alan Packard graduated from Colorado State University in 1978 with a PhD in Inorganic Chemistry. After receiving his PhD, he spent two years at the University of Cincinnati as a postdoctoral fellow studying technetium chemistry. In 1980, he joined the radiochemistry group of the Medical Department at BNL, and in 1982 he moved to Boston Children's Hospital/Harvard Medical School where he is currently a Senior Research Associate and Associate Professor of Radiology. His research interests include the*

*development of radiometal-labeled proteins for the diagnosis and treatment of disease and the development of  $^{18}\text{F}$ -labeled tracers for PET imaging of myocardial perfusion and neuroreceptors.*

Nuclear and radiochemistry have played and continue to play a critical role in nuclear medicine. From the study of nuclear reactions to produce isotopes for imaging and therapy to the integration of inorganic and organic chemistry and radiochemistry to produce radiopharmaceuticals to diagnose and treat diseases ranging from pediatric cancers to Alzheimer's Disease, nuclear medicine owes a great deal to both nuclear and radiochemistry. Nuclear medicine imaging is divided into two distinct subfields based in the type of emission (single photon or positron) that is being imaged. Planar and Single Photon Emission Tomography (SPECT) imaging use the photons emitted from a nucleus to create images of the distribution of radiolabeled compounds in the body. The radionuclides most often employed in single-photon imaging are  $^{99\text{m}}\text{Tc}$ ,  $^{111}\text{In}$ ,  $^{123}\text{I}$  and  $^{131}\text{I}$ .

The field of nuclear medicine owes much of its success to the development of the  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  generator at Brookhaven National Laboratory (BNL) in the late 1950s. Chemically, the  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  generator is amazingly simple. The parent radionuclide,  $^{99}\text{Mo}$  ( $t_{1/2} = 6$  d), is absorbed onto a small alumina column as  $^{99}\text{MoO}_4^{2-}$ , and the  $^{99\text{m}}\text{Tc}$  daughter ( $t_{1/2} = 6$  h) is eluted from the column as  $^{99}\text{TcO}_4^{1-}$  with normal (0.9%) sterile saline: a simple ion-exchange process. The 6 d half-life of  $^{99}\text{Mo}$  allows adequate time for the purification of the  $^{99}\text{Mo}$  parent, production and testing of the generators, and shipping to the end users, while the 6 h half-life of  $^{99\text{m}}\text{Tc}$  allows sufficient



time for preparation and quality control of the radiopharmaceutical before administration to the patient, localization of the compound within the patient, and image acquisition.

Interestingly, while  $^{99m}\text{Tc}$  is now used in more than 14 million procedures every year just in the US, the  $^{99}\text{Mo}/^{99m}\text{Tc}$  generator was never patented by BNL.



**Figure 1. The original BNL  $^{99}\text{Mo}/^{99m}\text{Tc}$  generator.**

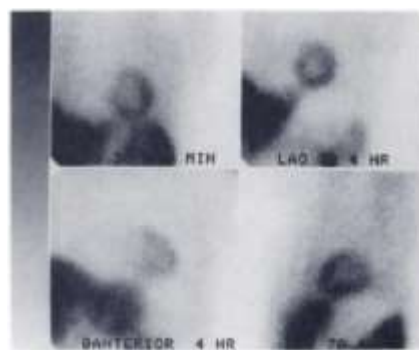
*Comments on patent application of Green, Richards and Tucker in 1958 for  $^{99}\text{Mo}/^{99m}\text{Tc}$  generator:*

*“While this method is probably novel, it appears the product will probably be used mostly for experimental purposes in the laboratory. On this basis no further patent action is believed warranted...” Atomic Energy Commission*

*“We are not aware of a potential market for  $^{99m}\text{Tc}$ ... We would recommend against filing...” Research Corporation for Associated Universities, Inc.*

Although the generator was developed in the late 1950s, there was a considerable time lag between its introduction and the development of the first practical method for the routine production of  $^{99m}\text{Tc}$  radiopharmaceuticals. This delay was due to the fact that  $^{99m}\text{TcO}_4^{1-}$  is not, in itself, a very useful imaging agent, except for imaging the thyroid and the gastric mucosa. To make agents that target other tissues, it's necessary to reduce the Tc from

the +7 oxidation state present in  $\text{TcO}_4^{1-}$  to +5 or lower where it can form complexes with ligands that can direct it to tissues of clinical interest. The first practical method to accomplish this was also developed at BNL, by Eckelman, Richards, and co-workers, who in 1971 reported the use of Sn(II) to reduce the  $^{99m}\text{TcO}_4^{1-}$  in the presence of DTPA to form a  $^{99m}\text{Tc}$ -DTPA complex for imaging the kidneys. This development, known as an “instant kit”, led to an explosion of new  $^{99m}\text{Tc}$ -based radiopharmaceuticals. When you consider these kits from the chemical point of view, it's really quite remarkable that you can add a saline solution of  $^{99m}\text{TcO}_4^{1-}$  to a vial containing Sn(II), a targeting ligand, and perhaps a buffer, shake it up, and “instantly” produce a sterile, pyrogen-free  $^{99m}\text{Tc}$  radiopharmaceutical in >95% yield and >95% radiochemical purity that is suitable for human use.



**Figure 2. First heart images obtained with a  $^{99m}\text{Tc}$ -isonitrile complex.**

Perhaps the most exciting subsequent advance in  $^{99m}\text{Tc}$  radiopharmaceuticals was the synthesis of the first effective  $^{99m}\text{Tc}$ -based myocardial perfusion agent by Davison, Jones, and co-workers in 1984<sup>8</sup>. This compound is itself quite interesting because, while the imaging agent is a Tc(I) hexakisisonitrile complex, it is synthesized in aqueous solution using a version of the original instant kit. One reason for the long delay between the development of the generator and the

<sup>8</sup> Holman, et al. A New Tc-99m-Labeled Myocardial Imaging Agent, Hexakis(t-Butylisonitrile)-Technetium(I) [Tc-99m TBI]: Initial Experience in the Human. J Nucl Med 1984 25:1350-1355.

development of the instant kit is that, in contrast to every other transition metal, there are no stable isotopes of Tc and therefore very little was known about the coordination chemistry of Tc in the 1960s. Technetium was first isolated in 1937 when Perrier and Segre identified it in a Mo cyclotron deflector from Lawrence Berkeley National Laboratory. It's interesting to note that in their description of Tc in third edition (1972) of "Advanced Inorganic Chemistry" Cotton and Wilkinson stated that "At the present time technetium has no uses, although the  $\text{TcO}_4^{1-}$  ion is said to be an excellent corrosion inhibitor for steels." Obviously, a lot has changed since then!

In contrast to single photon imaging, Position Emission Tomography (PET) is based on coincidence detection of the annihilation photons produced from the interaction of the positron that is emitted from the radionuclide with an electron in the body. The most common radionuclides employed in PET imaging are  $^{18}\text{F}$ ,  $^{11}\text{C}$ ,  $^{13}\text{N}$ , but there is increasing interest in a wide array of "non-traditional" positron-emitting radionuclides including  $^{64}\text{Cu}$ ,  $^{68}\text{Ga}$ ,  $^{89}\text{Zr}$ , and  $^{124}\text{I}$ .



Figure 3. Joanna Fowler preparing  $^{18}\text{F}$ [FDG] at BNL in the 1970s

The most frequently used PET imaging agent is  $^{18}\text{F}$ fluorodeoxyglucose ( $^{18}\text{F}$ [FDG]), a sugar analogue that is currently used for, among other things, the diagnosis of many types of malignant tumors. It was developed in the 1970s as a collaboration between Al Wolf's laboratory at BNL and the Reivich and Kuhl laboratories at the University of

Pennsylvania. The first human study with  $^{18}\text{F}$ [FDG] was carried out at the University of Pennsylvania using  $^{18}\text{F}$ [FDG] that was prepared at BNL and flown from Long Island to Philadelphia in small plane, which quite remarkable when you consider both the current regulatory environment regarding first-in-human use of radiopharmaceuticals and the 110 min half-life of  $^{18}\text{F}$ .

For cancer imaging,  $^{18}\text{F}$ [FDG] exploits the increased glucose uptake of many tumors (the Warburg effect) to visualize metabolic activity before and after treatment.

The FDA approval of  $^{18}\text{F}$ [FDG] in 1994 led to a dramatic increase in the number of cyclotrons in the United States and worldwide. It is estimated that there are close to 1000 cyclotrons installed worldwide solely for the production of medical radionuclides. With the increased availability of  $^{18}\text{F}$  and the emergence of other short-lived radionuclides for imaging, creative chemists have started to fill the toolbox of PET molecular imaging agents, including tracers for oncology, cardiology and neurology.

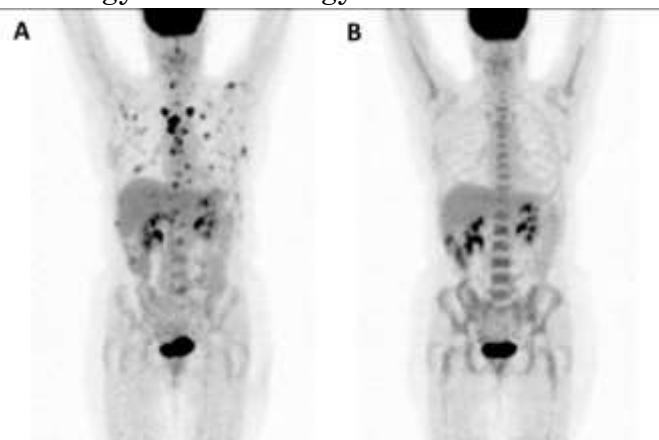


Figure 4.  $^{18}\text{F}$ fluorodeoxyglucose scan of a woman diagnosed with T-cell lymphoma. A. The image at diagnosis shows uptake in extensive disease sites along with normal signal in the brain and bladder, and B. The image following 4 months of chemotherapy shows the dramatic decrease in signal in the cancer sites indicating that this patient is responding well to therapy. (Image courtesy of Dr. Jonathan McConathy, Washington University in St. Louis.)

Recent years have seen PET make the transition from a new technology to an established method for non-invasive imaging. Its high sensitivity allows for investigation of biological processes at a cellular or molecular level without perturbing normal physiological functions. In addition to its uses in disease diagnosis and staging, PET imaging produces a map of drug distribution within the human body. Thus, it is invaluable for understanding pharmacokinetics (what the body does to a drug) and pharmacodynamics (what a drug does to the body).

In addition to small molecules labeled with  $^{18}\text{F}$  or  $^{11}\text{C}$ , new tracers are being developed based on peptides, antibodies, and nanoparticles labeled with  $^{68}\text{Ga}$ ,  $^{64}\text{Cu}$ , and  $^{89}\text{Zr}$ . Some of these compounds, such as antibodies, have very long blood circulation half-lives and thus require the use of longer lived isotopes (e.g.,  $^{89}\text{Zr}$ ,  $t_{1/2}=78.4\text{ h}$ ). New targetry, separation chemistry, and bioconjugation techniques are being developed which increase the availability of these isotopes and enables their translation to clinical studies. For example, Washington University in St. Louis and the University of Wisconsin - Madison both have a long history of production and distribution of these “non-standard” isotopes. Zirconium-89, in particular, has emerged as a useful radionuclide for monitoring the in vivo distribution of antibodies and antibody-drug conjugates. This strategy can be used to assess the distribution of newly developed drugs and also to stratify which patients might respond well to these highly targeted therapies.

In addition to imaging, therapy is also an important component of nuclear medicine. It is, in fact, the oldest component of the field with  $^{131}\text{I}$  being used to treat thyroid disease as early as 1948. Iodine-131 still plays an important role in treating thyroid diseases such as thyroid cancer and Graves' Disease. Other examples of radiotherapeutics include Bexxar ( $^{131}\text{I}$ ) and Zevalin ( $^{90}\text{Y}$ ), radiolabeled anti-CD20 antibodies used to treat non-Hodgkin's lymphoma, and Xofigo, the alpha emitter  $^{223}\text{Ra}$  that is used to treat metastatic prostate cancer.

Recently a combined strategy, known as theranostics, has emerged in which imaging is used to validate the therapeutic target paving the way for personalized medicine. In this strategy, an imaging radionuclide (e.g.,  $^{68}\text{Ga}$ ) is attached to a targeting vector such as an antibody, peptide or small molecule, and injected into the patient. After the presence of the target is verified by imaging, the study is repeated using a therapeutic radionuclide (e.g.,  $^{177}\text{Lu}$ ) that will localize in the target and kill the targeted tissue.

Moving forward, training the next generation of nuclear and radiochemists is vital to the ongoing success of this field: The development of new radiopharmaceuticals requires individuals who are well versed in a range of chemical specialties including nuclear and radiochemistry and also synthetic inorganic and organic chemistry. As the ACS's voice in nuclear and radiochemistry, NUCL has a vital role to play in ensuring the continued vitality of nuclear medicine and molecular imaging.

## In Memoriam: Alan Davison (1936-2015)



*Written by Liz McGrath, MIT Department of Chemistry*

Professor Emeritus Alan Davison, a fellow of the Royal Society who elevated the art of inorganic chemical synthesis in his laboratories at MIT for more than four decades, died Saturday, Nov. 14, at the age of 79 after a long illness.

Davison carried out his undergraduate studies in chemistry at the University of Swansea in Wales receiving a BS in 1959, followed by a PhD in 1962 in inorganic chemistry from Imperial College London under Nobel laureate Sir Geoffrey Wilkinson. Wilkinson was renowned for having pioneered inorganic chemistry and homogeneous transition metal catalysis.

After two years working as a chemistry instructor at Harvard University, Davison joined the Department of Chemistry at MIT in 1964 as an assistant professor in inorganic chemistry. By 1974, he had risen through the ranks of tenure to full professor.

"I first met Alan when he joined the MIT faculty where I was a second-year graduate student in Al Cotton's lab," says Stephen Lippard, the Arthur Amos Noyes Professor of

Chemistry. "He was a great source of practical knowledge, a man with a wonderful sense of humor, and a kind and patient mentor to me and many other graduate students. We maintained close contact over many decades, published together when I was at Columbia and then later at MIT, and remained friends for over five decades."

Throughout his illustrious career, Davison made research contributions spanning organometallic, boron, coordination, and bioinorganic chemistries. He was a serial innovator whose numerous seminal discoveries in diverse areas provided starting points for generations of followers. "His contributions to synthetic inorganic and medicinal chemistry were legion," Lippard adds. "He will be greatly missed."

"Alan was part of my life starting in 1967 and was involved in providing me with the opportunity to come to MIT in 1975," remarks Richard Schrock, the Frederick G. Keyes Professor of Chemistry. "He became my friend, colleague, and mentor."

Davison will be immortalized for his work with octahedral homoleptic isocyanide complexes of technetium(I). In collaboration with the late Professor Alun G. Jones, professor of radiology at Harvard Medical School, he brought rational synthetic understanding of technetium chemistry to the field of nuclear medicine. Davison and Jones' efforts included development of the first technetium-based myocardial perfusion-imaging agent, Cardiolite, an important tool in clinical nuclear cardiology.

"Alan Davison was an intuitive chemical genius who was gifted from an early age with magic hands in the laboratory," says Christopher Cummins, the Henry Dreyfus Professor of Chemistry. "He had a

photographic memory, a sharp wit, and an unusually deep understanding of people combined with compulsive empathy. He was also an amazing storyteller. This set of traits is seldom combined in a single individual, but their coexistence in the person of Alan Davison explains why the many students for whom he served selflessly as unofficial mentor were so drawn to him and so valued his wisdom and insights."

Former graduate student Chris Orvig PhD '81 remembers a particularly significant afternoon. It was the day he first met with Davison to be his teaching assistant, and Davison told him about his research project on the "new element" technetium. "I was hooked ... for life," Orvig says. "Alan's breadth of knowledge in chemistry and his unselfish devotion to graduate students (both his own and those of other faculty) were legendary. He always had time to encourage and mentor — I was privileged to share a few years with him," he says, adding, "I use the lessons of patience and understanding that I learned from him every day of my life."

"Alan Davison's mentorship had a profound effect on his many students and postdocs," says former Davison group member Mike Abrams PhD '83. "His brilliance and chemical insight matched with humor and compassion were a precious gift to all of us."

Throughout his illustrious career, Davison received many awards, including the Alfred P. Sloan Foundation Fellowship (1967); the Paul

C. Aebersold Award for Outstanding Achievement in Basic Science Applied to Nuclear Medicine (1993); the Ernest H. Swift Lectureship at Caltech (1999); the American Chemical Society Award for Creative Invention (2006); the Carothers Award for outstanding contributions and advances in industrial applications of Chemistry (2006); the Jacob Heskel Gabbay Award in Biotechnology and Medicine (2006); and the Society of Nuclear Medicine's Georg Charles de Hevesy Nuclear Pioneer Award (2009).

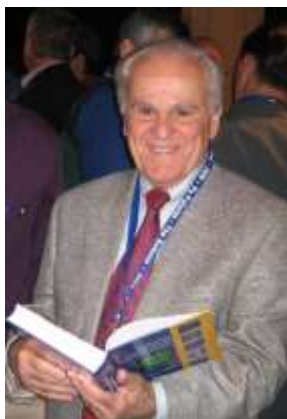
Former graduate student Jim Kronauge PhD '87 noted: "As well as having been an intuitive and enlightening chemist, Alan was also extremely modest. He never lobbied for any of the major awards he received, and did not think he had done anything special to deserve them. Alan was most proud of the students' teaching awards, and of course his admission to the Royal Society. Those accolades, he felt, were the most benevolent recognition by the people he loved."

On June 17, 2005, at a party held in his honor, attended by his many colleagues and former students, Davison was presented with a 20" x 20" bronze plaque bearing a description of his remarkable career. The plaque is mounted outside his old office on the 4th floor of Building 6.

Alan Davison is survived by his wife of 21 years, Lynne Davison of North Falmouth, Massachusetts; his children, Jackie, Fiona, Robert, Rowena and Ian; 16 grandchildren, and three great-grandchildren.



## In Memoriam: Gregory R. Choppin (1927–2015)



Co-discoverer of Element 101, mendelevium, Gregory R. Choppin, 87, passed away October 21, 2015, surrounded by his wife Ann and other family members. He was born November 7, 1927 in Eagle Lake, Texas. Greg's undergraduate studies at Loyola University Chicago were interrupted by service in the U.S. Army during the post-WW2 occupation of Japan. Greg returned to study chemistry at the University of Texas, earning a Ph.D. before completing a post-doc and working as a research scientist at the University of California Berkeley with Dr. Glenn Seaborg. After his work at the Lawrence Radiation Laboratory, Greg became a faculty member at Florida State University, where he fostered the careers of students through his teaching and research. Greg had received many awards recognizing his exemplary teaching and research contributions, and FSU had previously created a named chair in chemistry in his honor. For more, please see the obituary published in the [Tallahassee Democrat](#).

---

*Remembrances from Ken Nash, Ph.D., Florida State University, 1978, under the supervision of Professor Greg Choppin:*

"I first met Greg a few days after Christmas in 1971 when, during the winter break at my undergraduate institution, I fled a Chicago

snowstorm with a couple of my fellow Lewis College colleagues to meet Professor Choppin and to see FSU as a possible grad school destination. Though I freely admit that the 72°F temperature, sunshine, and balmy breezes made a profoundly positive impression, it was clear from our first brief contact that Greg would be an exceptional mentor for the pursuit of a Ph.D. in chemistry. Greg's easy-going manner was evident, but it was also obvious that he had a clear focus on conducting world-class science and a joy in discovery that was infectious. During the years of my graduate education, indeed throughout his career, this enthusiasm was always evident."

"Greg also exhibited great patience with my process of self-discovery, which included a "my-choice" M.S. and a year in exile learning that I was not an industrial R&D chemist. After completing my Ph.D., I was fortunate enough to eke out a career in f-element science, which meant that I was often in contact with Greg, with his research, and with many generations of "Choppinites", a fraternity of former students, postdocs, and other visitors to Greg's labs. Greg always sought to maintain a close and cordial connection to this group of his "mentees". As a result there were generally several opportunities for impromptu (and occasionally planned and organized) gatherings of the clan almost every year. Each and every such event was always memorable."

"In closing, I must plead "guilty" to occasionally re-orienting the 35 mm slides that he used for presentations (long after most of his colleagues had moved on to transparencies), but I didn't originate the concept of tinkering with Greg's slides. After a while, I only did it to have Greg refer to me as one of those "smawrt asses" who play with his slides. Professor Choppin was (and still is

through the values he instilled in his minions)  
an exceptional mentor of young scientists, and  
though we will miss his ever present smile, we

will never forget his life lessons. Thanks  
Greg.”