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Exploring the Frontiers of Nuclear and Radiochemistry

Darleane C. Hoffman*

Graduate School, Department of Chemistry, University of California, Berkeley and Nuclear Science Division, MSR0319, Lawrence Berkeley National Laboratory, Berkeley, CA 94720-8169, USA

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Frontiers in fundamental research on both nuclear and chemical properties and prospects for future advances were highlighted in this two-day Symposium. The following four areas were especially emphasized: Superheavy elements — production, chemical and nuclear properties, and potential use of the unique techniques developed to solve applied problems; Nuclear processes as chemical probes to explore environmental and ecological radionuclide distributions; Application of nuclear and radiochemical techniques to geochemical, pharmaceutical, and biological sciences, and even information technology; Environmental radiochemistry and fundamental actinide sciences and practical applications in nuclear waste storage and remediation. These topics were explored in both invited lectures and poster presentations. The Global Nuclear Energy Program recently proposed by the U. S. is briefly described and the potential need for scientists with expertise in nuclear and radiochemistry and actinide science, and the opportunities for synergistic interactions are highlighted.

1. Recognition of Prof. Hiromichi Nakahara's 70th Birthday Year

Prior to my plenary talk, long-time friend and colleague, Professor Hiromichi Nakahara, was congratulated on his 70th birthday year. He has been professor and/or mentor to many of the Symposium participants and a tireless advocate for the importance of education and research in this field and has been especially active in promoting international collaborations.

Prof. Nakahara received his Ph. D. from Massachusetts Institute of Technology (MIT) in 1966 in the group of Charles D. Coryell, J. Irvine, Jr., and Glenn Gordon**,** but remained in the Boston area for about 8 months working at Cambridge Nuclear, a company which sold radioisotopes. Concurrently, he studied nuclear physics with Prof. Grodinz in MIT Physics Department.

In 1967, he returned to Japan and in July was employed by Prof. T. Nishi as an Assistant in the Engineering Research Institute of Kyoto University. In 1971, he went to the Inorganic Chemistry Group of Niigata University as Assoc. Professor and in 1975 joined the nuclear chemistry group of Prof. Y. Murakami at Tokyo Metropolitan University and was promoted to full professor in 1981.

He retired in early 2000 at the age of 63 and has suffered some major health problems, but has managed to overcome these, and recently took on the heavy responsibility of organizing the Symposium for the 50th Anniversary Celebration of Nuclear and Radiochemistry in Japan sponsored by the Japan Society of Nuclear and Radiochemical Sciences (JNRS). This was held in conjunction with its 2006 Annual Meeting which began earlier this week on October 24 in Mito, Japan and extends through October 27. Prof. Nakahara was instrumental in the founding of the JNRS which has been so important to students and teachers and in strengthening Nuclear and Radiochemistry programs in Japan.

I was in Berkeley in 1978–79 on a Guggenheim Fellowship just prior to returning to Los Alamos to be Division Leader and I still traveled often to Berkeley. Prof. Nakahara and his first student Prof. Hisaaki Kudo became well acquainted with Prof. Glenn Seaborg in the early 1980s when then postdoctoral student Kudo joined Prof. Seaborg's group. I am not sure when I first met Prof. Nakahara, but I came to know him and nuclear and radiochemistry in Japan very well after he successfully proposed me for a Japan Society for the Promotion of Science (JSPS) lectureship, and I spent nearly 4 weeks lecturing and visiting various universities, institutes, historic and cultural sites and museums throughout Japan.

2. Introduction

Prof. Y. Hatano, Director of the Advanced Science Research Center (ASRC), gave an insightful introduction to the Symposium in which he emphasized the necessity for continued support for research in the field of Nuclear and Radiochemistry and its continuance in the curriculum of institutions of higher education in Japan and worldwide.

Dr. Yuichiro Nagame deserves our heartiest congratulations for his vision in proposing and organizing this "forward-looking" and extremely stimulating Symposium emphasizing the many and diverse Frontiers of Nuclear and Radiochemistry, including novel developments and latest achievements.

In this two-day Symposium we will broaden our horizons by exploring the many Frontiers of Nuclear and Radiochemistry that are open before us. One of the perceived problems with the field of nuclear and radiochemistry is that it is difficult to define precisely in the same way that we might define organic chemistry, biochemistry, inorganic chemistry, analytical chemistry, or geochemistry, for example. Our expertise is applicable to so many different areas that we sometimes lose our identity — the science community tends to forget who we are and the necessity for fostering the training and education that allow us to see new opportunities and make seminal contributions to an amazingly wide spectrum of science and applied science.

Included in the Symposium are invited lectures and posters organized around the topics: A. Superheavy elements — production, and chemical and nuclear properties; B. Nuclear processes as chemical probes; C. Application of nuclear and radiochemical techniques; D. Environmental radiochemistry and actinide sciences. There was time for considerable discussion and comment after the talks. An impressive technical tour of J-PARC was also conducted for the participants.

*Corresponding author. E-mail: dchoffman@lbl.gov. Fax: +1-510-486-4818.

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3. Session A

In the first session on Superheavy Elements (SHEs) – production, and chemical and nuclear properties, the latest reports of production of isotopes attributed to new elements, the systematics of production cross sections, studies of mechanisms of production reactions, chemical properties of the heaviest elements, and plans for new systems and techniques for studies of both nuclear and chemical properties were presented.

These are all extremely difficult experiments, at the furthest reaches of the Chart of the Nuclides and the Periodic Table, with cross sections dipping below the picobarn level. Thus, long running times and highly radioactive targets are often required, making independent confirmation of results at different laboratories nearly impossible in some cases. However, over time, the knowledge base to make connections with known systems can be carefully constructed in order to verify reported results using other reactions as evidenced by the results of Morita et al., Paper SE04.

Much progress has been made, even in this difficult field over the last 10 years, as can be illustrated by looking at the status of knowledge about production reactions and nuclear and chemical properties of the heaviest elements described in my 1997 talk on "Frontiers of Heavy Element Nuclear and Radiochemistry" given at the Asian-Pacific Symposium On Radiochemistry (APSORC-97).[1] In the periodic table of mid-1997 shown in Figure 1,[1] the Transactinides or TANs were highlighted. (TANs include all the elements from 104 and on up as far as we can reach, and so include the Super Heavy Elements (SHEs), regardless of exactly how they may be defined.) Although elements had been reported through 112, no element beyond Meitnerium (109) had yet been validated by IUPAC/IUPAP so elements beyond Meitnerium had not been named. Since that time, Nielsbohrium (Ns) for element 107 has been changed to Bohrium (Bh) and Hahnium (Ha) for element 105 has been changed to Dubnium (Db). Currently, all of the elements through 118, with the exception of 117, have been reported but not yet confirmed.

A "renaissance" of interest in studies of the chemical properties began in the late 1980's in order to compare properties of the TANs with their lighter homologues in groups 4, 5, and 6 of the periodic table. This effort was sparked by predictions that relativistic effects might result in deviations in chemical properties from expected trends within these groups; chemical studies were facilitated by the production of longer-lived isotopes of these elements, especially of seaborgium (element 106). New instrumentation was developed for both gas-phase and solution chemistry and attempts to use the SISAK (Special Isotopes Studied by the AKufe technique) system for rapid chemical studies were initiated.

It was suggested that ^{269,270}Hs might be produced using ^{26}Mg projectiles with ^{248}Cm targets and a definitive study of this reaction has been published recently by Dvorak et al.[2] Isotopes of elements from 113 through 118 (with the exception of 117) have been reported by Oganessian (SE02) and Oganessian et al.[3–5] One of the problems is that these chains do not end with previously known isotopes. Morita et al. (SE04), using the ^{209}Bi(^{70}Zn,n) reaction have observed evidence for 2 decay chains of element 113 with mass 278 which decay to known isotopes of dubnium. This is a different mass number from those of 283 and 284 reported by Oganessian et al.[5] for element 113 as daughter products of ^{287,288}115 produced via ^{243}Am(^{48}Ca,*x*n) reactions, and if confirmed, would constitute an independent claim to production of element 113 via a direct reaction. All of these reports await confirmation.

Figure 2 shows a Periodic Table as of early 2006. Note that the elements 110 and 111 are now officially named Darmstadtium (Ds) and Roentgenium (111). A Joint Working Party (JWP) of the IUPAC/IUPAP will soon review claims to first production and positive identification of the elements heavier than 111 to determine whether the additional evidence for element 112 is sufficient to assign priority of discovery to the GSI group and ask them to propose a name. The JWP may also consider claims to discovery of still heavier elements. There are many different versions of the chart of the TAN nuclides and several are shown in this Symposium.

A contour plot is given in Figure 3. It shows some of the "stepping stones" of shorter nuclides produced on the way to the region of stability predicted in the late 1960s and 1970s to exist near the doubly magic spherical region centered around Z = 110 to 112 and N ~ 184. Half-lives as long as a billion years were originally predicted, but none were ever "discovered" and recent theoretical calculations have drastically reduced the predicted half-lives.

The Berkeley Gas-filled Separator (BGS) for "pre-separation" of the desired species prior to chemical and/or nuclear studies and the successful use of SISAK with BGS as a pre-separator are described by Nitsche et al. (SE03). The TransActinide Separator and Chemistry Apparatus (TASCA) currently being tested at GSI and the opportunities for a variety of studies there are described by Schädel in this Symposium (SE02).

We heard from speakers from many countries — Russia, Germany, U. S., Japan, China, Belgium, Portugal, Switzerland,

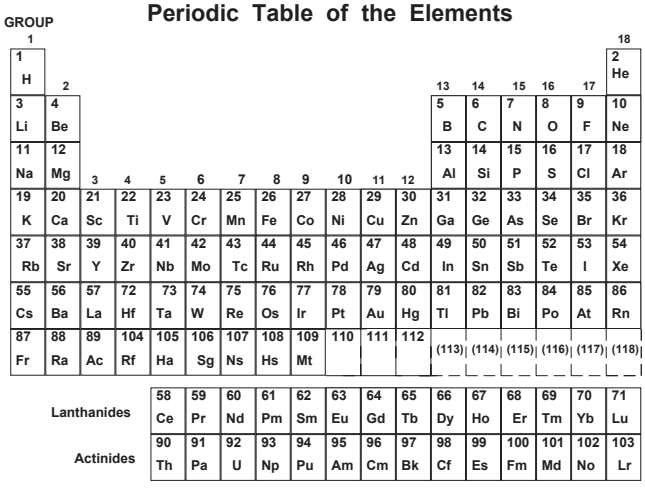

Figure 1. 1997 Periodic Table.[1]

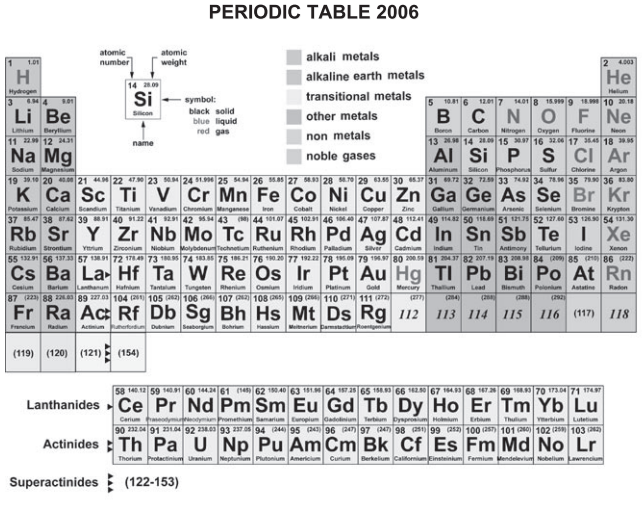

Figure 2. 2006 Periodic Table. Elements in italics have been reported but not yet confirmed.

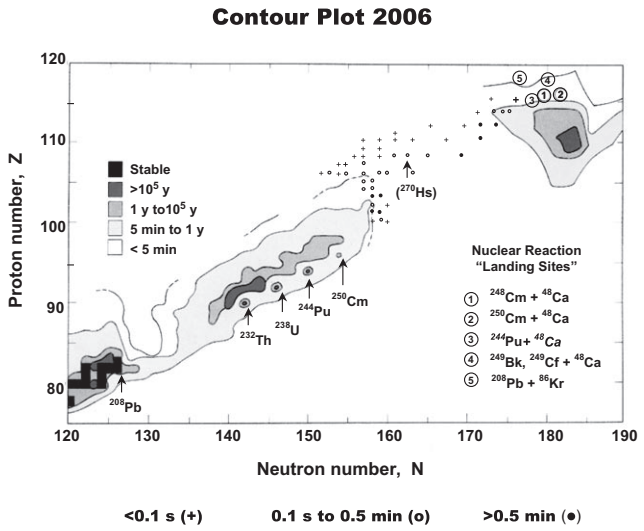

Figure 3. Contour Plot 2006.

and of course, many are involved in large international collaborations.

SHE research is an area that really requires federal support in the U. S. as it cannot easily be incorporated within most funded applied programs and has not found support within NSF (National Science Foundation) or the DOE Office of Science. Some funding may become available under the newly initiated GNEP (Global Nuclear Energy Partnership) program but it would be more focused on Actinides than TANs although some of the equipment developed might be applicable such as ultrasensitive analysis for forensic applications.

We have often pointed out to the funding agencies that there is a severe current and future shortage of nuclear and radiochemists, especially actinide chemists, and that the exotic, frontier SHE studies attract many undergraduate and graduate students to nuclear and radiochemistry and provide excellent education and training for applied as well as fundamental research in relevant areas such as: Ultrasensitive and radioanalytical analyses; Surveillance of clandestine nuclear activities and nuclear forensics; Automated, computer-controlled remote processing systems; Nuclear medicine, isotope production, radiopharmaceutical preparation, and diagnostics and therapy; Nuclear power reactor design and performance; Treatment, processing, and minimization of nuclear waste; Nuclear waste isolation and site remediation; Environmental studies including prediction and monitoring of behavior of actinides and other species in the environment.

4. Sessions B, C, and D

Now this leads me rather directly into the use of Nuclear Processes as Chemical Probes described in **Session B.** Applications to both exotic research topics such as changing the electron-capture decay rate of ^{7}Be by placement inside C_{60} cages to frontier measurements using time-differential perturbed angular correlation (TDPAC) to measure electromagnetic fields, and the use of short-lived electron-capture emitters for strain measurements in nanometer layers were discussed.

Session C on the Application of Nuclear and Radiochemical Techniques is an especially "rich" session including talks on applications ranging from uses in radiopharmaceuticals, nanotechnology, nanosafety, and even information technology and cognitive science to geochemical characterization of the Oklo natural reactors predicted by Kuroda, the development of RIMS for ultra-trace level analysis and determination of isotope ratios of long-lived nuclides applicable both in exotic research and environmental studies, and accelerator based mass spectrometry measurements of ^{36}Cl for reassessment of atomic bomb radiation dosimetry. This was a truly far-ranging and impressive session.

And last, but not least, in **Session D** on Environmental Radiochemistry and Actinide Science, sensitive ion mass spectrometry was used to show that transport of plutonium on submicrometer particles was responsible for its long-distance transport in the ground water from the Mayak Production Association, Russia. Studies of tritium in the atmosphere, radioecology of I, and microbial interactions with Pu in the environment were discussed.

5. Future?

Some promising signs have been seen in the U. S. There is a growing recognition that there is a shortage of nuclear and radiochemists and the National Nuclear Security Agency has implemented funding of University Programs to help train the next generation of researchers in low energy nuclear science with expertise relevant to the U. S. national security missions in both energy and defense.

The public is beginning to realize that nuclear energy is the best solution to the "Greenhouse Effect" problem and avoids other undesirable impacts on the environment associated with most other energy sources. For example, even Patrick Moore, an avid environmentalist and co-founder of Greenpeace, now makes the case for nuclear energy in his article, "Nuclear Re-Think", IAEA Bulletin 48/1, September 2006, pp. 56–58.

Perhaps one of the most encouraging signs was that President George W. Bush announced plans for a new Global Nuclear Energy Partnership (GNEP), and a revitalization of U. S. Nuclear Energy Programs. In a radio address on February 18, 2006, President Bush stated, "This morning, I want to speak to you about one part of this initiative: our plans to expand the use of safe and clean nuclear power. Nuclear power generates large amounts of low-cost electricity without emitting air pollution or greenhouse gases." He further said that "…my Administration has announced a bold new proposal called the Global Nuclear Energy Partnership. Under this partnership, America will work with nations that have advanced civilian nuclear energy programs, such as France, Japan, and Russia. Together, we will develop and deploy innovative, advanced reactors and new methods to recycle spent nuclear fuel. This will allow us to produce more energy, while dramatically reducing the amount of nuclear waste and eliminating the nuclear byproducts that unstable regimes or terrorists could use to make weapons." The GNEP proposes a global solution to issues that need to be addressed in the current global nuclear environment and would entail a policy change to favor recycling by "fuel cycle" states in order to manage the long-term nuclear waste problem, and concurrently reduce proliferation risks. Spent fuel would be viewed not as a waste product, but as an asset to be managed to provide needed nuclear power.

To help support the research and development required for this initiative, the U. S. DOE Office of Basic Energy Sciences sponsored a workshop called "Frontiers in Chemical Research for Advanced Nuclear Energy Systems" in early August 2006. The charge to the Workshop was to identify basic research needs and opportunities in advanced nuclear energy systems and related areas, with a focus on new, emerging and scientifically challenging areas that have the potential to have significant impact on science and technologies. The following panels were convened: Materials under extreme conditions; Chemistry under extreme conditions; Separations science; Advanced actinide fuels; Advanced waste forms; Predictive modeling and simulation; Crosscutting and grand-challenge science themes. A full report is in preparation. It is apparent that the expertise of nuclear and radiochemists, especially actinide chemists will be much in demand if these areas are to be successfully

addressed.

To ensure that there is a future for nuclear and radiochemistry, we must all do a better job of communication, both domestically and internationally. We must continue to emphasize the need for education and training in Nuclear and Radiochemistry to our colleagues in Chemistry and Physics and other sciences, as well as with the general public, the funding agencies, and governments! We must build on the educational and training programs we have and increase their visibility and capacity in order to sustain and enhance our capabilities. **New frontiers are ahead!**

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