PROFESSOR ADAM SOBICZEWSKI: SEMINAL CONTRIBUTIONS TO UNDERSTANDING AND PREDICTING NUCLEAR STABILITY OF THE HEAVIEST ELEMENTS

Darleane C. Hoffman

Department of Chemistry, University of California, Berkeley
Seaborg Institute for Transactinium Science and Nuclear Science Division
MS 70R0319, Lawrence Berkeley National Laboratory
Berkeley, CA 94720-8169, USA

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Dedicated to Adam Sobiczewski in honour of his 70th birthday

Throughout his long career Adam Sobiczewski has made major contributions to the interpretation and understanding of experimental results on the synthesis and decay properties of the heaviest nuclei and to the development of theoretical methods for predicting properties of as yet undiscovered nuclei. He led the way in using a dynamical approach to macroscopic-microscopic calculations of spontaneous fission half-lives of deformed nuclei in a multi-dimensional deformation space without adjustable parameters. Examination of the well-deformed even-even nuclei with Z=104–114 and N=142–176 showed that inclusion of higher order deformations greatly increased the N=162 shell correction. Nuclei in the region of the doubly magic deformed nucleus $^{270}108$ were predicted to be much more stable than previously believed and significant deviations from the old rule of rapidly decreasing spontaneous-fission half-lives with increasing Z should occur. The impact and influence of these results on experimental investigations and the interactions between theory and experiment will be discussed.

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1. Introduction

I first met Adam Sobiczewski when he and V. M. Strutinsky came to visit us at Los Alamos National Laboratory in November 1969. I had met Strutinsky, then at the I. V. Kurchatov Institute of Atomic Energy in Moscow, at the August 1966 International Symposium on *Why and how should we investigate Nuclides far off the Stability Line* held in Lysekil, Sweden. On behalf of the Los Alamos Radiochemistry Group, I had reported on the production of heavy elements in a recent Los Alamos thermonuclear test detonated underground at the Nevada Test Site in May 1966. We were puzzled because we failed to find nuclides any heavier than ^{257}Fm even though our yield extrapolations indicated they should have been produced and our crude extrapolations of half-lives indicated that they were long enough that we should have detected them. So we were desperately searching for explanations about both the cross sections for successive neutron captures in ^{238}U and for predictions of the expected stabilities of the still heavier elements which we believed should have been produced. Strutinsky and many others reported on their calculations of heavier nuclides, including superheavy elements. However, I was particularly impressed with his shell corrections (based on Nilsson's levels of the oscillator shells) to the liquid drop model and how well his calculations fit the measured masses in the actinide region. Minima in the potential energy plots as a function of deformation that might be related to the fission isomers observed in some of the elements from Th to Cm were also seen. I was fascinated with the calculations for several as yet undiscovered even Z elements including 104, 108, 112, 116, and had Strutinsky's detailed Russian article on *Shells in Deformed Nuclei* translated for our use at Los Alamos and continued some contact with him concerning his calculations.

In 1969, the Welch Foundation convened a Conference on *The Transuranium Elements: The Mendeleev Centennial* in Houston, Texas on November 17–19. Adam Sobiczewski and Strutinsky were invited to attend as Round Table Discussion Leaders and were granted permission to visit us in Los Alamos prior to the meeting. They were among the first visitors to Los Alamos from what were then called the "Eastern Block countries" and in those "Cold War" days, the extent of their travel was rather narrowly proscribed, but we managed to show them some of the spectacular scenery and nearby pueblos of northern New Mexico.

The 1969 Welch Conference [1] was a memorable one as there were many fast-breaking results, many new ideas, new experiments on the discovery of element 104, new calculations of superheavy element half-lives and other properties, and just reported observations and theoretical interpretations of fission isomers. It provided a wonderful forum for the exchange of ideas among scientists from all over the world. After this initial meeting, I saw Adam Sobiczewski at subsequent conferences and read many of his papers. I came to especially respect him for his clear and concise presentations of insightful and carefully formulated concepts and his equally thorough calculational approaches, which took account of as many as possible of the most important factors determining nuclear stability. He was not content to devise a model and then just make a myriad of small changes in the various parameters but attempted to focus on the most important considerations. He began his career in the field of prediction of the nuclear stabilities of actinide and SuperHeavy Elements (SHEs) during the time of the big wave of interest in SHEs sparked in the late 1960s and 1970s by predictions of an "island" of stability well beyond uranium around magic numbers of 126 or 114 protons and 184 neutrons. After receiving his Ph. D. he worked at the Joint Institute for Nuclear Research in Dubna and then returned to the Institute for Nuclear Research in Warsaw. He was a member of the Berkeley-Lund-Warsaw group that gave the first extensive and realistic estimates of the stability and SF half-lives of SHEs on the basis of the modified-oscillator model [2,3]. Later calculations [4,5] indicated that element 110 with 184 neutrons should be the longest-lived with a half-life in the range of ten thousand to a billion years.

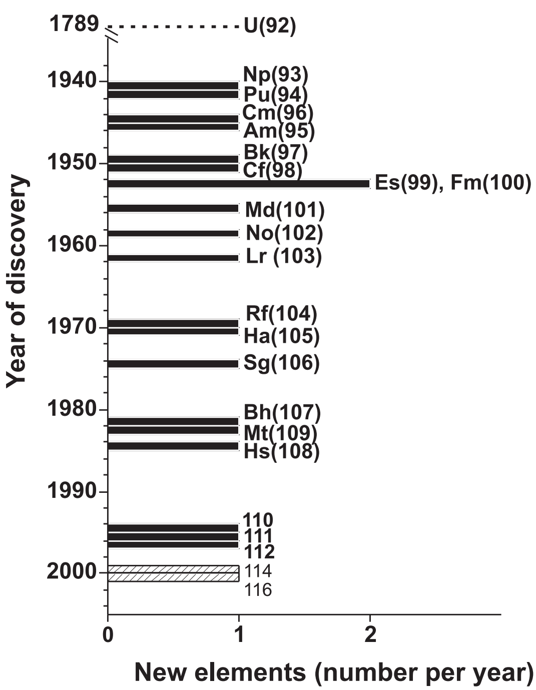

Fig. 1. Timeline of discovery of transuranium elements.

However, the wave of theoretical and experimental interest began to wane in the 1980s when early reports of discoveries of SHEs in nature and at accelerators could not be confirmed. By the end of 1987 no credible evidence remained for SHEs, either in nature or artificially produced and most researchers abandoned the quest. But Adam Sobiczewski and his group continued research in this area and continued to make major contributions to our understanding of the stability of the heaviest elements. By 1984 the elements through 109 had been reported as shown in the timeline of the discoveries of the transuranium elements in Fig. 1. After an interval of some ten years, elements 110 through 112 were reported between 1994 and 1996. These discoveries initiated a renaissance of interest in experimental searches for SHEs and a number of experimental groups built improved on-line separator systems to look for still heavier elements. Evidence for elements 114 and 116 has been reported but has not yet been confirmed.

2. Predictions of spontaneous-fission half-lives and nuclear stability

In 1991, Patyk and Sobiczewski [6, 7] reported that macroscopic–microscopic calculations which included larger deformation spaces showed that nuclides in the region of the deformed shells at 108 protons and 162 neutrons should be much more stable toward SF than previously believed. The N=162 shell correction was greatly increased by the inclusion of higher order deformations and was larger than that for the known 152 neutron shell. This was calculated to lead to large increases in stability for the deformed nuclei near the doubly magic deformed nucleus ^{270}Hs. The shell-stabilized nuclei in this region were called "deformed superheavy" nuclei. It was further predicted that the SF half-life for Sg with 162 neutrons would be longer than that for any isotope of element 104. Because of the large shell effects, strong deviations from the rule that SF half-lives decrease rapidly with increasing atomic number should occur.

This was of particular interest to nuclear chemists who were planning the first chemical studies of Sg using 0.9-s ^{261}Sg, then the longest known Sg isotope. These predictions motivated researchers to try to produce longer-lived Sg nuclides and at the Actinides-93 Conference, the discovery of ^{265,266}Sg with half-lives of 2 to 30 s and 10 to 30 s, respectively, were reported [8], thus helping to substantiate this important theoretical breakthrough.

In 1995, Sobiczewski together with Smolańczuk and Skalski [9] published a widely quoted, landmark article entitled, *Spontaneous-fission half-lives of deformed superheavy nuclei*. It summarized some of the results presented earlier and described a dynamical approach to the calculation of SF half-lives of deformed nuclei in the region of the doubly magic deformed nucleus ^{270}Hs in a multidimensional space without any adjustable parameters. (In older papers, a two-dimensional deformation space that included only deformations of the lowest multipolarities was used.) Even-even nuclei with Z=104 through 114 and N=142 through 176 were considered. These were chosen because they were all expected to be well deformed and a sufficiently large deformation space was required to be able to calculate even the ground-state energies, but the fission barriers were relatively simple and thin compared to those with Z=92 through 102, and, finally, most of these nuclei would not exist at all if it were not for shell effects. Thus this put stringent requirements on the theory to properly account for these effects. In addition, there were some experimental measurements of fission half-lives in this region with which the theoretical calculations could be compared. A four dimensional space appropriate for these heavy nuclei was used which disregarded the odd-multipolarity deformations important for the lighter nuclei with more complex fission barriers, but included the higher multipolarities β_6 and β_8 which are important for describing the shell effects for these "deformed superheavy" nuclei. Results of these calculations for the SF half-lives together with the alpha-decay half-lives and the experimentally determined values as of mid-1997 are shown in Fig. 2. It can be seen that the fit between the experimental and predicted values is exceedingly good and these predictions of half-lives have served as a guide for experimentalists in planning experiments to produce and study nuclides in this region for many years.

Muntian, Patyk, and Sobiczewski [10] recalculated properties of even-even nuclei with Z=82–128 and N=126–188 using more recent data for masses in the optimization of the macroscopic part of the energy, and a different method for adjusting the pairing force strength in the microscopic part than in their previous calculations in order to assess the sensitivity of the various calculated properties to these modifications. They found that the recalculated values of alpha-decay energies and half-lives were quite close to their previous calculations, lending credibility to the predictions.

Calculations of the collective properties of the deformed nuclei in the region of the doubly magic deformed nucleus ^{270}Hs have also been performed by Muntian and Sobiczewski [11]. The energies of the first 2^+ excited states and the branching ratios between alpha-decay of a nuclide to the first 2^+ state and the ground state of its daughter were given for even-even nuclides with Z=102–112 and N=146–166. Recently, calculations of collective properties such as deformations and rotational energies have been extended to even-even nuclei with Z=82–126 and N=126–190 and the influence of the deformations of various multipolarities on the rotational energies was considered [12]. They suggested that experimental measurements of these properties could be used to deduce whether the extent of deformation is as large as predicted for ^{270}Hs and how fast the deformation may change from one nuclide to another — again excellent guidance for the experimentalist.

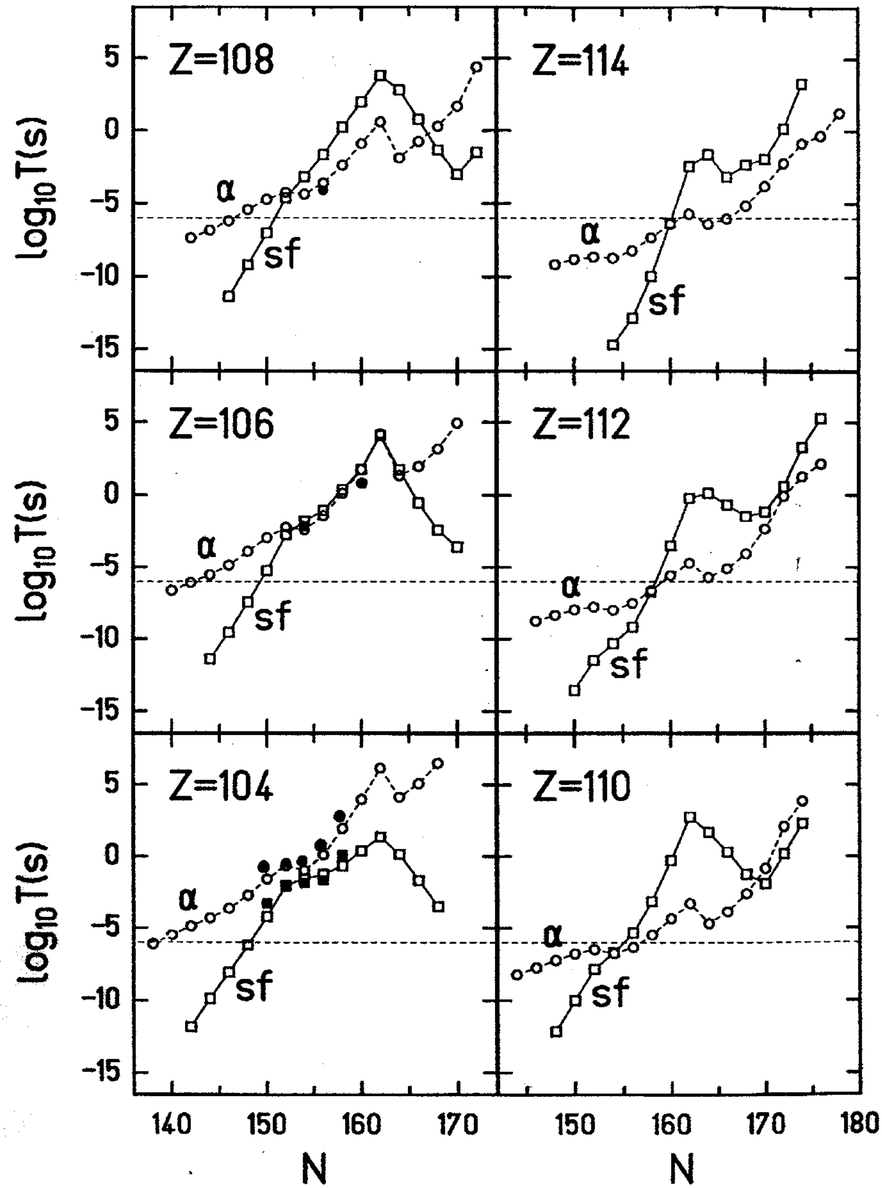

Fig. 2. Experimental partial half-lives for SF and alpha-decay of even-even isotopes (solid symbols) as of 1997 plotted together with the theoretical predictions for even-even isotopes of elements 104–114 from Ref. [9].

3. Interactions between theory and experiment

As mentioned earlier, because of the prediction of extra stability for isotopes nearer N=162 in the region of Z=108, the successful experiments were undertaken in which the longer-lived isotopes ^{265,266}Sg were produced. These isotopes were long enough for chemical studies and subsequently were used by an international group working at GSI, Darmstadt, Germany to perform the first chemical studies of Sg (106) in both the gas-phase and in solution [13,14]. These experiments showed that Sg behaved similarly to the periodic table group 6 elements Mo and W rather than to U(VI) and many additional studies of the properties of Sg have been performed.

Nuclear chemists were encouraged by this experimental support for the predictions of extra stability for isotopes closer to the N=162 deformed shell, and experiments were undertaken at the 88-inch cyclotron at the Lawrence Berkeley National Laboratory to try to produce ^{267,266}Bh (107) using ^{249}Bk(^{22}Ne, $4n$, $5n$) reactions. From predictions of the SF and alpha half-lives for the even-even nuclides similar to those shown in Fig. 2, half-lives of the order of 10 s were expected. Indeed, ^{267}Bh with a half-life of $\sim$17 s and a production cross section of about 70 pb was identified based on its alpha decay to known daughter isotopes [15]. Its SF branch could not be determined due to the SF background from Fm produced by transfer reactions. No positive identification of ^{266}Bh could be made although SF systematics based on known SF half-lives suggest that its SF half-life should be relatively long because of the odd-proton and odd-neutron hindrance factors [16] and it should decay by alpha emission. The first chemical studies of Bh were proposed by Gäggeler *et al.* and scheduled at the cyclotron at the Paul Scherrer Institut in Switzerland even before the discovery of the longer-lived Bh isotope, illustrating the confidence in the predicted stability due to the proximity to the deformed shells. Experiments ^{267}Bh with an on-line isothermal gas chromatography system showed that a volatile oxychloride of ^{267}Bh was formed as expected for a group 7 element, but that it was less volatile than those of its lighter homologues Tc and Re [17].

Experiments with rotating ^{248}Cm targets and ^{26}Mg beams have been conducted at the UNILAC accelerator at GSI and the chemical separation of seven atoms of ^{270,269}Hs as the volatile tetroxide has been reported [18]. A novel cryogenic gas adsorption chromatography column was used which also served as the detection system for identification of ^{270,269}Hs based on detection of alpha-decay to known Sg and Rf daughter isotopes. The experiments showed that Hs formed a very volatile oxide similar to that of Os tetroxide measured in the same system and provides qualitative evidence that Hs is also a member of group 8 of the periodic table. The decay characteristics of three of the Hs decays agreed well with those of ^{269}Hs, previously observed in the alpha-decay chain of $^{277}112$ [19]. Two decay chains were incomplete and could not be definitely assigned. Two of the observed Hs decays were attributed to ^{270}Hs, but its half-life cannot be measured directly in this system and was deduced to be about 4 s based on the observed alpha-decay energies. In order to compare with the predictions that it is a doubly magic deformed nucleus, it is important to try to obtain a more precise measurement of the half-life of ^{270}Hs, perhaps by a direct measurement and also to determine if it has a SF branch.

Experiments are planned at LBNL to use the Berkeley Gas-filled Separator (BGS) to separate $^{271}110$ produced via the ^{238}U(^{37}Cl,4n) reaction and identify it based on its alpha-decay to known isotopes. The alpha-decay energy of ∼10 MeV estimated by interpolation between predicted masses for adjacent even isotopes would lead to half-lives of only tens of ms, but due to the odd nucleon, longer half-lives might be expected. Nuclides with half-lives as short as 100 microseconds can be separated and identified with the BGS, but if longer-lived nuclides can be produced then chemical studies might be performed after using BGS as a pre-separator.

All of the isotopes reported as of mid-2001 for elements from Sg through element 116 are shown in Fig. 3. It should be noted that the isotopes of elements 114 and 116 and their alpha-decay daughters await confirmation by other groups.

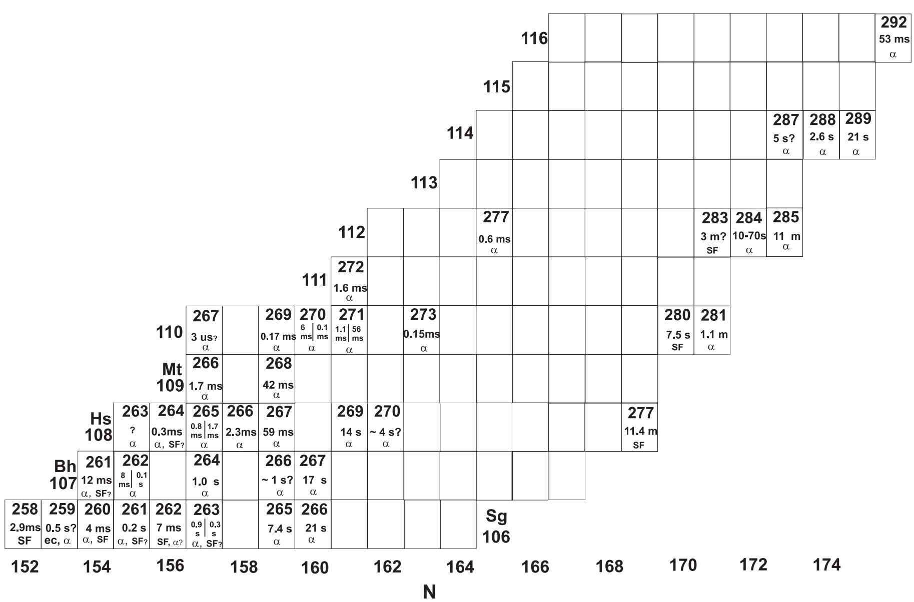

Fig. 3. Isotopes of Sg through element 116 reported as of mid-2002.

4. Conclusion

Sobiczewski presented [20] a short review of the theoretical studies based on the macroscopic-microscopic description in which he pointed out that the shell effects in deformed superheavy nuclei are large and comparable to the shell effects calculated for the spherical superheavy nuclei. He contended that the existence of nuclei in the doubly magic deformed region around $Z=108$ and $N=162$ has changed our previous view of the stability of the heaviest nuclei. No longer are the spherical SHEs around the hypothetical doubly magic spherical $^{298}114$ nucleus expected to form an island in the "ocean" of total instability, but rather they belong to the extended "usual" peninsula of relatively long-lived nuclides. Thus, we may expect that nuclei all along the way to the region of spherical superheavy nuclei can be observed provided we can find ways to synthesize them. Many nuclei in the superheavy region will decay predominantly by alpha-emission which should make their experimental identification much easier and more certain. I have attempted to illustrate this concept (Fig. 4) by plotting the nuclides discovered between 1978 and mid-2002 as a function of Z and N on a contour plot of uncertain origin (originally in color) dating back to at least 1978 that was included in a Science article on Superheavy Elements [21].

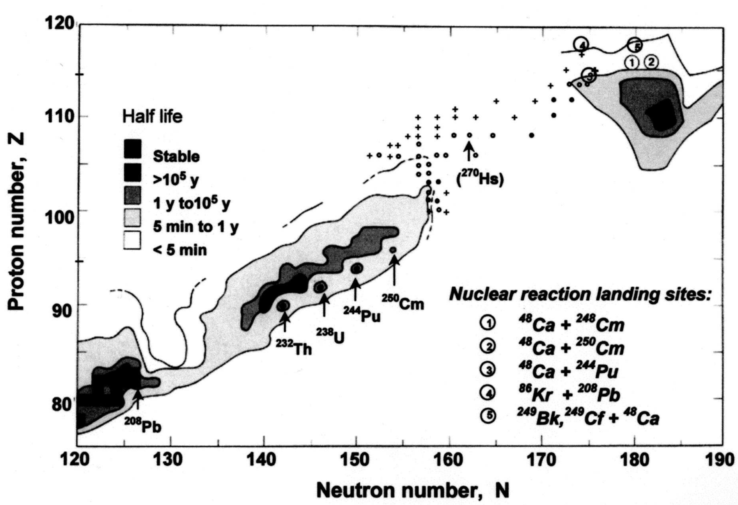

Fig. 4. Plot of heavy element topology from ∼1978 showing heavy element isotopes reported from 1978 up to mid-2002 with symbols denoting the following half-life ranges: 0.1 ms to 0.1 s (+); 0.1 s to 5 min (∘); > 5 min (•). The doubly magic deformed nuclide ^{270}Hs is indicated. Landing sites for some proposed production reactions for spherical SHEs are shown.

Adam Sobiczewski was invited to present the 1999 J.M. Nitschke Memorial Lecture at LBNL. In his address, entitled *Structure and Properties of Superheavy Nuclei*, he discussed the current status of both theoretical and experimental advances in the field. New data obtained in the past few years tend to support his concept that nuclei with half-lives long enough to be observed will exist all the way from the region of deformed SHEs to the region of spherical SHEs. A photo of him taken as he delivered his lecture is shown in Fig. 5 together with a poster announcing the event. We wish him many more years of fruitful research in nuclear theory and anticipate he will continue his synergistic interactions with experimentalists in the endeavor to gain an ever more complete understanding of nuclear stability and its limits.

I wish to express my gratitude to Jørgen Randrup for many helpful comments and his gracious assistance in rendering this manuscript into LATEX format with which many of us "older" experimentalists are not conversant.

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