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Current Status and Future Prospects on Super Heavy Nuclei Research (Summary of the Symposium)

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Current Status

During the SHE 2015 meeting, all aspects of one of the key questions in *Nuclear Physics*, **the synthesis and properties of Super Heavy Elements (SHE)** were discussed. Since 1981, remarkable progress has been made in synthesis of new heavy and super-heavy nuclei (SHN). Six new elements ($Z = 107$ to 112) have been produced and identified in ‘cold’ fusion reactions and six ($Z = 113$ to 118) in ‘hot’ fusion reactions. The nuclei were produced at accelerators delivering beams of heavy ions from calcium to zinc at intensities of about $1 \text{ p}\mu\text{A}$ ($1 \text{ p}\mu\text{A} = 6.24 \times 10^{12}$ particles/s). In-flight separation is now commonly used for element discovery and for pre-separation for chemistry experiments. The new nuclei were efficiently separated from the beam by vacuum or gas-filled recoil separators. Position sensitive detector systems were used for identification and measurement of the decay properties. The experiments were sensitive enough to measure cross-sections as low as 0.02 pb for synthesis of $^{278}113$ in the reaction $^{70}\text{Zn} + ^{209}\text{Bi}$. However, the experiments also revealed an increase of cross-sections up to 8.5 pb for synthesis of $^{288}115$ in the reaction $^{48}\text{Ca} + ^{243}\text{Am}$, the latter increase attributed to the approach of the compound nuclei to a predicted island of stability at $Z=114$ and $N=184$. Measured lifetimes for alpha decaying nuclei demonstrated increases in stability for nuclei located in an island of deformed nuclei at proton number 108 and neutron number 162 and again in the region of $Z = 114$.

The Uniqueness of the Heaviest Elements

Because of large atomic numbers, superheavy atoms and nuclei differ significantly from lighter species; this presents unusual challenges for nuclear and atomic theory. As the very existence of superheavy nuclei hinges on a dynamical competition between short-range nuclear attraction and huge long-range Coulomb repulsion, precise calculations are difficult. Nuclear theory at the limits of mass and charge involves large extrapolations; hence, it is crucial to estimate errors on computed observables.

There exists a general consensus among current theoretical models with respect to general properties of superheavy nuclei: neutron and proton separation energies, shapes, and energies of emitted alpha particles but no consistent predictions of lifetimes exist. The main source of uncertainty is fission. Unfortunately, current models of fission are not robust, both in terms of modeling and input, and associated uncertainties remain difficult to assess. While theory consistently predicts that stability will increase with the addition of neutrons in these systems as one approaches $N = 184$ the exact location of center of the island of stability, is still an open question. Some theoretical predictions identify $Z = 114$,

$N=184$ as a doubly closed shell spherical nucleus. Others favor $Z=120$, $N=184$ or $Z=126$, $N=184$. In superheavy nuclei, the density of single-particle energy levels is fairly large, so small energy shifts, such as the regions of enhanced shell stabilization in the superheavy region near $N = 184$, are generally expected to be fairly broad; that is, the notion of magic numbers and the energy gaps associated with them becomes fluid there. New experiments planned for synthesizing additional isotopes of element 118 and the new element 120 will shed light on the strength of the predicted shells or subshells at $Z=114$, 120 or 126 and $N=184$. As the region of the transfermiums is explored, it is expected that the shell model will be subjected to sensitive tests. In this respect, quality spectroscopic data on excited states of superheavy nuclei are essential to constrain theory.

Paths to the Heaviest Elements

From the experimental point of view, pushing the limit for the discovery of SHE up to $Z=118$ in fusion reactions was made possible due to the tremendous progress in increasing the intensity and reliability of stable light and medium mass heavy ion beams at Dubna and GSI, up to $2 \mu\text{A}$ for ^{48}Ca beams. Even so, recent attempts by Dubna and GSI to produce elements $Z=119$ and 120 SHE indicated upper limits in cross-sections around 60 fb for these elements.

Actinide targets have been used to produce nine of the fifteen superheavy elements synthesized to date, including elements 104-106 and 113-118. Virtually all research on superheavy nuclei currently underway or in the planning stage involves the use of actinide targets, principally ^{237}Np , $^{239,240,242,244}\text{Pu}$, ^{243}Am , $^{245,248}\text{Cm}$, ^{249}Bk , and $^{249-251}\text{Cf}$ (mixed isotope target). While some of these isotopes are currently available in the required amounts, ^{244}Pu , ^{248}Cm , ^{249}Bk , and mixed Cf are in limited supply or require special production and processing. New trans-actinide targets, notably long-lived Cf isotopes recovered from spent ^{252}Cf sources will offer new opportunities for heavy isotope production. Facilities for actinide production and processing, including the High Flux Isotope Reactor and Radiochemical Engineering Development Center at Oak Ridge National Laboratory (ORNL) were described.

Fusion reactions continue to occupy center stage in the efforts to create new elements. Although several phenomenological models appear capable of calculating heavy isotope production cross sections, a closer investigation of these reveals that they differ significantly in their individual components. It was suggested that part of this was due to compensating effects associated with different mass predictions in the heavy element region. Uncertainties in fission barriers have very large effects on the cross section calculations (as well as on the determination of half-lives and decay modes) and the ability to determine barriers more accurately was called into question. Indeed new theoretical investigations of the fission process raise further questions about the adequacy of calculations based on fission barriers and the assumption of adiabatic evolution on the potential energy surface. Superheavy elements are expected to be formed at the final stages of the r-process nucleosynthesis, especially in strong r-process scenarios involving neutron star mergers. Here, the challenge is to explain fission of very neutron-rich superheavy nuclei.

New investigations of the projectile dependence of fusion cross sections indicate smaller residue cross sections for projectiles more massive than ^{48}Ca . Further experiments are needed to clarify the relative importance of neutron richness or neutron skins and of structural effects. Careful explorations of quasi-fission systematics are serving to better quantify the contact times and capture probabilities as a function of entrance channel mass asymmetry. The development of predictive models for such small production cross sections, which are capable of guiding experimental efforts remains an important challenge for theoretical efforts. Even the best models available are not able to predict the production cross sections of SHE in hitherto unexplored reactions or regions with sufficient reliability.

In addition to its critical importance in addressing the main topic of this workshop, the production of and properties of, relatively stable superheavy elements, a clear understanding of reaction mechanisms will also facilitate the exploration of transient systems with extremely large Coulomb effects. Exotic density distributions such as bubbles and toroids have been predicted for such nuclei but no convincing experimental evidence for these structures has yet been obtained.

The possible use of new beams of neutron rich radioactive isotopes was extensively discussed. While access to new neutron rich isotopes in the trans-actinide region should be possible in favorable cases, it was generally agreed that intensity limitations of the new generation of rare beam accelerators will limit the possibility of new element searches at these facilities in the near term.

On the more positive side, impressive progress has been made in microscopic TDHF calculations using interactions derived from density functional theory to explore the reactions relevant to new isotope production. Results from these calculations are quite consistent with recent experimental investigations comparing fusion cross sections for different projectiles.

Though still relatively unexplored, multinucleon transfer reactions involving heavy partners offer the possibility of overcoming the N/Z limits imposed by the fusion reactions. They provide an important alternative method to search for yet unknown neutron rich heavy and superheavy isotopes. Results from studies of $^{136}\text{Xe} + ^{208}\text{Pb}$, used to test theoretical predictions, are encouraging for giving additional insight into the stability in the vicinity of the superheavy island.

Chemical Studies

In the superheavy element region the large Coulomb charges lead to strongly attracted electrons moving at considerable fractions of the speed of light. Such velocities cause relativistic mass increases and thus, a contraction of the orbital radius of these electrons occurs. Due to this contraction and the resulting shielding of the nuclear charge, other electrons with higher angular momentum (e.g. d-electrons) become more weakly bound and their orbital radii expand. These effects dramatically influence the chemical properties of such elements. Chemical studies of the heaviest elements with atomic numbers $Z \geq 101$ provide not only crucial and challenging opportunities to advance our understanding of properties of matter at the limits of existence but also to elucidate the influence of relativistic effects on atomic electrons and the structure of the Periodic Table of Elements (PTE) at its farthest limit. Determining the atomic properties of the heaviest elements, such as electronic configurations, ionization potentials, atomic/ionic radii and redox (reduction and oxidation) potentials can provide important information on electronic structure influenced by strong relativistic effects.

Spin-orbit mixing and electron correlation effects make the theoretical description of chemical properties here extremely complicated. Thus, experimental results elucidating these chemical properties are required for benchmarking and further developing the existing models.

As both half-lives and production rates of these nuclides are rapidly decreasing with increased atomic number, they are usually available in quantities of only a few atoms or often one atom at a time. Thus, the experiments are very challenging. So far, the chemistry of SHE with atomic numbers 104-108, i.e. elements rutherfordium, dubnium, seaborgium, bohrium, hassium, and elements 112-114, i.e. copernicium, ununtrium, and flerovium has been investigated in at least one experimental campaign. These challenging experiments revealed that SHE behave chemically close to their lighter homologues in the corresponding groups (i.e. columns) of the PTE, generally following established trends. However, the increasing relativistic effects are clearly observed in these first-glimpse experiments, particularly for the elements copernicium and flerovium, members of groups of the PTE showing increasing metallic character with increasing atomic number Z , which revealed extraordinary volatilities. The Fl experiment using TASCA (TransActinide Separator and Chemistry Apparatus) at

GSI revealed that Fl is a volatile metal, the least reactive element in group 14, but not as inert as a noble gas. This behavior is in good agreement with recent fully relativistic calculations.

Recently, a new electrochemical approach based on an atom-at-a-time scale has been developed by the JAEA group. They conducted redox experiments of the heaviest elements, oxidation of nobelium (No) and reduction of mendelevium (Md) using a newly developed flow electrolytic column chromatographic method. In addition, they successfully measured the first ionization potential (IP_1) of the heaviest actinide element.

Investigations of the formation and detection of the volatile seaborgium (Sg) compound $Sg(CO)_6$ were conducted with cryo-thermochromatography coupled to the recoil separator GARIS (Gas-filled Recoil Ion Separator) at RIKEN, Japan. The result demonstrated that the short-lived atoms (^{265}Sg , half-life $T_{1/2} \sim 10$ s) still form a chemical compound in single-atom quantities.

Future Prospects

New Facilities

Significant increases in beam intensity would greatly aid future programs of superheavy element research. Towards that goal new high current injectors and accelerators are under construction. The most powerful new facility will be the ‘SHE Factory’ under construction at FLNR in Dubna. The DC280 cyclotron will provide beams of medium mass heavy ion projectiles ($A = 20-60$) with energies up to 10 MeV/u and intensities of 5–10 μA . With beam intensities up to 10 μA , low cross-section experiments can be performed in shorter time, cross-section limits can be pushed into the region of 1 fb, and physical studies of SHN can be performed, which are presently not possible.

Also under construction at GANIL (Caen) is the SPIRAL2 accelerator complex, based on high power low energy superconducting LINAC able to deliver up to 5 kW of medium mass heavy ion beams (10–20 μA of ^{48}Ca for example). Another heavy ion RFQ injector is foreseen at SPIRAL2 for higher ion masses $A > 60$ up to U beams. A new generation of Spectrometer–Separator, with high rejection power and excellent mass resolution at the focal plane, S3 under construction, will be coupled to the high intensity heavy ion beams of SPIRAL2. An advanced demonstrator for a superconducting CW-Linac is under development at GSI for the future replacement of the UNILAC machine at GSI with the same goal of higher beam intensities. In addition design of the next generation of Super-Conducting separators and innovative detection systems have also started both at RIKEN/GARIS, GSI, DUBNA, and S3 at GANIL.

While intensities will be limited, new RIBs or upgrade of existing facilities like SPIRAL2 at GANIL, TAMU at Texas A&M, or ReA3 at FRIB (USA) have launched research studies on fusion reactions and massive mass transfer processes using neutron rich ‘exotic beams’. These facilities may contribute to the exploration of new neutron rich heavy species. With the upcoming second-generation Rare-Isotope (RI) facilities the discussion started to which extent RI beams can contribute to SHE research. Problems are low beam intensities and limited beam-time. First generation experiments are reaction studies including fusion-fission, fast fission, and transfer. Such studies are possible with a minimum beam intensity of $10^6/s$ available at FRIB, SPIRAL2, HIE-Isolde, and FAIR over long isotopic chains. Key questions are the choice of the best reaction and target-projectile combinations to approach the $N=184$ neutron shell and to go beyond element 118. The influence of isospin, deformation, and shell structure in target and projectile and in the compound nucleus, respectively, is not understood at present. The new generation RI facilities will deliver an intensity of the order of $10^9/s$ for rare isotopes

near the stability line to produce isotopes in the region from uranium to rutherfordium for direct mass measurements and in the future possibly for in-beam spectroscopy.

New Instrumentation

In association with these new accelerator facilities, it is mandatory that new instrumentation, capable of using these facilities as effectively as possible, be developed. This process has already begun. Next generation instruments with optimized separation efficiency have gone into operation at RIKEN and are under development at JINR Dubna and at Texas A&M. Vacuum separators such as velocity filters or recoil mass separators are being used in a complementary fashion. New developments are under way at JINR Dubna and at SPIRAL2 with the S³separator.

The performance of recoil separators should be improved. High transmission and high suppression of the background are mandatory. In experiments using this modernized instrumentation further open questions related to SHN will be investigated. Among these is the extension of the island of stability as function of proton and neutron number, the study of shape and spin isomers, the investigation of both neutron rich and neutron deficient fragments from quasi-elastic and quasi-fission reactions.

A new generation of broad acceptance spectrometers designed to deal with the wide range of isotopes, energies and angles produced in such reactions are required. The higher intensity beams and new target technologies to become available at the Dubna Superheavy Element Factory and other facilities (e.g. SPIRAL2 and S3) should be of great utility in this emerging area. Alternate techniques, which greatly reduce the lifetime limitations imposed by spectrometers, should also be pursued as predicted half-lives for the heaviest elements fall well below spectrometer transit times.

Ion catcher systems in combination with magnetic separation, ion traps, or Multi-Reflection Time-Of Flight (MRTOF) spectrometers open up new perspectives for direct identification of SHE, high-precision mass spectrometry, precise decay spectroscopy and optical spectroscopy of heavy elements up to the rutherfordium region. With the new stable-beam SHE factories and improved efficiency their application will be extended to heavier species. While the mother-daughter correlation method commonly used at present is limited to α emitters and their decay chains to known daughter nuclei, the direct identification can be applied to all nuclear species independent from their decay mode and being sensitive for a wide range of lifetimes. Research topics include mapping the shell landscape and fission barriers of SHE starting from uranium, passing the transition region, from liquid drop to shell nuclei, and finally explore the nature and extension of the predicted spherical region of SHE. With laser spectroscopy in the first step relativistic effects on the electrons of the heaviest elements are explored. Such experiment is under way at GSI for nobelium. Stopping of the separated reaction products in gas catchers and injection of the low energy beams into penning-trap or MRTOF spectrometers would allow for isobaric purification, accurate mass determination and. Compared to present techniques, long half-lives as expected for some of the neutron rich SHN, are not a limitation, on the contrary, the precision increases with increasing half-life. In addition, atomic beam experiments such as e.g. collinear laser spectroscopy and Stern-Gerlach experiments will also become possible.

Chemical Investigations

The third generation of experimental chemistry campaigns with superheavy elements (SHE) is approaching. This opportunity is induced by the design and construction of new even more powerful particle accelerators compared to the existing one and by the experiment durations possible at these accelerator complexes running specifically for superheavy element research only. Indeed, this is required since the SHE are artificially produced in heavy ion induced nuclear fusion reactions at single-

atom-at-a-time quantities and they are known to have limited half-lives mainly in the order of seconds and milliseconds.

Chemical investigations of SHE need improved, faster, more sensitive, and if possible more efficient experimental techniques to chemically identify even heavier SHE and to get a deeper look into the chemistry of the lighter SHE. The following experimental approaches are promising for future research on the chemical properties of the heaviest elements:

In gas phase:

- Metal organic chemistry assessing low oxidation states of SHE (Sg – Mt);
- Reaction gas chromatographic systems with reactive stationary surfaces (Cn – 118);
- Fast vacuum adsorption chromatographic systems for elemental states (Cn – 118);
- Stern Gerlach experiments, Laser Ionization experiments (113,115);
- Mass trap / chemistry in the trap experiments (Db – 118).

In liquid phase:

- Electrochemistry for noble transition metals for redox potential (Hs – Rg);
- Crown ether/ionic liquids extraction for ionic radii determination (Db – 115);
- Ion exchange chromatography for complexation chemistry and hydrolysis (Db – Rg).

The advocated new generation facilities will require:

- The development of innovative target technologies able to stand the mechanical, thermal, and charging stress connected to highest beam intensities;
- The availability of rare actinide target material such as e.g. $^{242,244}\text{Pu}$, ^{243}Am , ^{248}Cm , ^{249}Bk , $^{249-251}\text{Cf}$, ^{254}Es ;
- The design and construction of highly efficient physical pre-separators dedicated for chemistry.

For all these investigations high quality theoretical calculations are pivotal.

Production and Fabrication of Targets, in particular Actinide Targets

The Production and Fabrication of Actinide Targets session reviewed the production and chemical processing of heavy actinide materials, target fabrication techniques, and projected actinide target needs for near term research on superheavy nuclei.

In advance of the session, projections for target needs through 2021 were requested from the Joint Institute for Nuclear Research (Russia), RIKEN (Japan), Lawrence Berkeley National Laboratory (USA), GANIL (France), Argonne National Laboratory (USA), and GSI (Germany). These projections were updated at the workshop and shared with the broader community. They provide a snapshot of planned superheavy element research over the next five years, including synthesis of heavy isotopes of element 118, searches for elements 119 and 120, and advances in SHE spectroscopy and understanding of nuclear structure and reaction mechanisms.

The session included an overview of actinide production, target fabrication, and material availabilities. The impact of actinide materials in super-heavy element discovery was reviewed, and strategies for enhancing the production of rare actinides including ^{249}Bk , ^{251}Cf , and ^{254}Es were described. Facilities for actinide production and processing, including the High Flux Isotope Reactor and Radiochemical Engineering Development Center at Oak Ridge National Laboratory (ORNL), and the PIK High Flux Reactor, currently under construction at the St. Petersburg Nuclear Physics Institute, were described.

Due to the extremely high beam doses required for the synthesis of heavy and superheavy nuclei, production and fabrication of targets has always been a challenging issue. With a significant increase in beam intensity target resistance against thermal and radioactive damages may become a limiting factor.

Further R & D for the improvement of targets is a critical task. Presentations from GANIL, GSI, the University of Mainz and ORNL addressed specific issues and progress in fabrication, characterization, and production of stable and radioactive actinide materials and targets.