Results and Perspectives for the Study of Heavy and Super-Heavy Nuclei and Elements at IMP/CAS (Postprint)

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Abstract

In the past 30 years, synthesis of new isotopes and study of their decay properties have been remaining as high-priority research program at the Institute of Modern Physics, Chinese Academy of Sciences (IMP/CAS). Up to now, 34 new isotopes have been synthesized, 22 of which are heavy and super-heavy nuclei. In this paper, the techniques used and the results obtained at IMP/CAS for the study of heavy and super-heavy nuclei and elements are reviewed chronologically and the perspectives in the near future are introduced.

Full Text

Preamble

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Abstract

Over the past 30 years, the synthesis of new isotopes and study of their decay properties have remained a high-priority research program at the Institute of Modern Physics, Chinese Academy of Sciences (IMP/CAS). To date, 34 new isotopes have been synthesized, 22 of which are heavy and super-heavy nuclei. This paper reviews the techniques used and results obtained at IMP/CAS for the study of heavy and super-heavy nuclei and elements in chronological order, and introduces future perspectives for this research.

Keywords: α decay; Fusion and fusion-fission reactions; Super-heavy nuclei

[Figure 1: see original paper] Overview of heavy and super-heavy nuclei studied at IMP/CAS

1 Introduction

Driven by human curiosity about the unknown, the synthesis and discovery of new elements to expand the Periodic Table has become one of the frontier research fields in nuclear physics and chemistry. Over the past 70 years, laboratories worldwide have made significant efforts to synthesize heavy and super-heavy nuclei/elements and study their physical and chemical properties. In China, such studies began in the 1990s. Since then, numerous new isotopes have been synthesized for the first time and their decay properties studied experimentally, establishing this as a high-priority research program at the Institute of Modern Physics, Chinese Academy of Sciences (IMP/CAS). To date, 34 new isotopes have been synthesized, 22 of which are heavy and super-heavy nuclei. Figure 1 shows the location of these isotopes in the chart of nuclides.

Chronologically, these studies at IMP/CAS can be divided into three stages. During the early stage, isotopes of interest were separated using radiochemical methods and their decay properties studied via γ rays or β-γ/X-γ coincidences. Subsequently, the helium-jet technique was developed and used to separate and transport products away from reaction sites, enabling detection of their subsequent α, β and/or γ decays. Beginning in 2010, a new gas-filled recoil separator was commissioned, allowing fusion-evaporation residues to be transported and separated, with the energy-position-time correlation method employed to assign charge number Z and mass number A of the isotopes of interest.

This paper reviews the techniques and results obtained at IMP/CAS chronologically in Sections 2–4, and introduces future perspectives for the study of heavy and super-heavy nuclei and elements in Section 5.

2 The First Stage

During the first stage, studies focused primarily on neutron-rich heavy nuclei. Isotopes of interest were produced via multi-nucleon transfer reactions

or neutron-induced reactions, separated using radiochemical methods, and their decay properties studied through γ-rays or β-γ/X-γ coincidences.

2.1 208,209Hg

The neutron-rich isotopes 203,205–209Hg were synthesized and their decay properties studied by bombarding thick natural Pb targets with 12C or 18O beams at HIRFL (Heavy Ion Research Facility in Lanzhou) at IMP/CAS, leading to the first synthesis of new isotopes 208Hg and 209Hg. In the study of 208Hg [1, 2], high-efficiency release, separation, and collection of Hg products was successfully achieved with good selectivity using a special offline gas-phase thermochromatographic process followed by liquid-liquid extraction. Elemental assignment of the mercury isotope was based on identification of its β-decay daughter Tl observed in periodically extracted Tl element samples growing in the separated Hg product solution. The half-life of 208Hg was measured to be $42^{+23}{}_{12}$ min. Later, the device was upgraded for online use in synthesizing the 209Hg isotope [3]. Six neutron-rich mercury isotopes 203,205–209Hg were observed using β-γ coincidence measurement techniques. The half-life of 209Hg was determined to be $35^{+9}{}_{6}$ s, with four γ rays following its β-decays assigned. The half-life of 208Hg was remeasured and improved to $41^{+5}{}_{4}$ min.

2.2 185,186Hf, 237,238Th and 239Pa

Experiments searching for new isotopes 185Hf [2, 4] and 237Th [5] were carried out at the 300 kV Cockcroft-Walton accelerator at Lanzhou University and the 600 kV machine at IMP/CAS using 14 MeV neutrons produced by deuteron reactions with a rotating TiT (tritide titanium) target. Samples containing approximately 20 g of natural metallic tungsten powder and 16 g of $(NH_4)_2U_2O_7$ powder were irradiated in the searches for 185Hf and 237Th, respectively. Experiments for the other three new isotopes [6–8] were performed at HIRFL, with nuclei produced via intermediate-energy heavy ion (18O) bombardments of thick natural uranium or tungsten targets through multi-nucleon transfer reactions.

After bombardment, irradiated targets were rapidly transferred to a chemical laboratory via a pneumatic transport system and dissolved in hydrochloric or nitric acid. Products of interest were then separated radiochemically from the mixture of reaction products using different processes, and sources were prepared for γ counting by HPGe detectors. Assignments were primarily based on observation of growth and decay of characteristic γ rays from daughter nuclei. The half-lives of 185,186Hf, 237,238Th and 239Pa were determined to be $3.5\$\pm0.6min, 2.6\pm1.2min, 5.0\pm0.9min, 9.4\pm2.0min, and 106\pm\$30$ min, respectively.

2.3 175Er and 197Os

Experiments searching for new isotopes 175Er [9] and 197Os [10] were carried out at the 600 kV Cockcroft-Walton accelerator at IMP/CAS using 14 MeV

neutrons, as described previously. Several natural ytterbium targets and platinum metal foils were used to produce 175Er and 197Os via (n,2p) reactions, respectively. A rabbit system quickly transported irradiated samples into a well-shielded lead chamber. HPGe planar detectors and two coaxial HPGe detectors, or a clover detector consisting of four n-type coaxial germanium detectors, were used for X-ray and γ-ray measurements, respectively. Energy and time spectra for X and γ rays were recorded in both singles and coincidence modes. Identification was made through coincidence between characteristic X and γ rays. The half-lives of 175Er and 197Os were determined to be $1.2\$\pm0.3min and 2.8\pm0.6min, respectively, with partial decay schemes proposed based on X-\$$ and γ-γ coincidence measurements.

3 The Second Stage and Helium-Jet Period

During the second stage, the helium-jet technique was developed at IMP/CAS and extensively used to transport products away from reaction sites, reducing background essential for measurements of neutron-deficient isotopes and heavy and super-heavy nuclei.

3.1 Experimental Setups and Detection

Beams of 22Ne, 26Mg, 32S, 36Ar, or 40Ca with energies of 5–7 MeV/amu from HIRFL entered a target chamber filled with ~1.1 atm helium gas, passed through a thick Havar window, and bombarded self-supporting metal targets such as 58Ni, 92Mo, 96Ru, 106Cd, 112Sn, or 241,243Am. Reaction products recoiling out of the target foil were thermalized in helium gas and attached to aerosol (NaCl, KCl or $PbCl_2$) clusters. Helium jets then swept these clusters and reaction products through a Teflon capillary, implanting them into a rotating wheel, movable tape, or tantalum disk in a collection chamber.

Depending on specific experiments, collected samples were processed using different procedures. In studies of heavy and super-heavy isotopes, samples were rotated away from the deposition site and positioned in front of a series of Si(Au) surface-barrier detectors for periodic α particle detection. Precursors were identified using correlations between detected α decay energies and times. In studies of light and medium-mass neutron-deficient isotopes, a tape station system was employed. After collection, radioactivity on the tape was moved to a shielded counting chamber for coincidence measurements of proton, γ and/or X-ray energies and times, while the next sample was being collected.

Two fully depleted silicon surface barrier detectors (300 µm thick) for proton measurements were located on opposite sides of the movable tape, with a coaxial HPGe detector behind each silicon detector for measuring γ(X) rays. Precursors were identified through correlations between β-delayed protons and γ rays from the $2^+ \rightarrow 0^+$ states of the daughter nucleus. When X rays were observed with good statistics, assignment was made via X-γ coincidences.

3.2 Super-Heavy Nuclei: 258,259Db and 264,265,266Bh

259Db [11] and 258Db were produced via the reaction 241Am(22Ne, 4-5n) at Elab=118 MeV, while 266Bh [12], 265Bh [13] and 264Bh were produced via 243Am(26Mg, 3-5n) at Elab=162 and 168 MeV. Reaction products were transported and collected using the helium-jet technique and rotating wheel apparatus. α-decays of products and their daughter nuclides were detected by a set of Si(Au) detectors. The Z and A of nuclides were unambiguously identified through genetic relationships between new activities and known nuclides established by recoil-α coincidence measurements.

The new nuclide 259Db was determined to have a half-life of 0.51$±$0.16 s and decays by α-particle emission with Eα=9.47 MeV. The nuclide 258Db has a half-life of 4.3$±$1.1 s and decays by α-particle emission with Eα = 9.08, 9.17 and 9.30 MeV, in agreement with previously known data [14], thus confirming the reliability of our 259Db assignment.

A total of 8 correlated decay events of the new isotope 265Bh and 4 decay events of the known isotope 264Bh were observed. 265Bh decays with a half-life of $0.94^{+0}\cdot{}^{70}{}_{0\cdot31}$ s by emission of α particles with an average energy of 9.24$±$0.05 MeV. For 266Bh, 4 correlated decay events were observed. The measured α-particle energy and half-life for 266Bh are $9.03\$\pm0.08 MeV and 0.66^{\{+0\}\cdot}\{59\}\_\{0\}.\{26\}\$$ s, respectively. The Eα value is close to the 9.07 MeV observed for 266Bh in the first chain of element 113 at RIKEN, supporting the elemental assignment of element 113 [15].

The isothermal gas chromatographic behavior of group 5 elements Nb, Ta and Db was also investigated [16] in a brominating atmosphere using the OLGA (On-Line Gas chromatography Apparatus) technique. It was found that Db forms a very volatile compound, most likely the pentabromide, which is more volatile than similar compounds formed under identical conditions with Nb and Ta.

3.3 Light and Medium-Mass Neutron-Deficient Nuclei

Using the p-γ coincidence technique in combination with the helium-jet tape transport system, β-delayed proton decays of nine new nuclides in the rare-earth region near the proton drip line (121Ce [17], 125Nd [18], 128Pm [18], 129Sm [18], 135Gd [19], 137Gd [18], 139Dy [18], 142Ho [20], and 149Yb [21]) and five nuclides in the A=90 region with N Z (81Zr [22-24], 85Mo [22-24], 89Ru [25], 92Rh [26], and 93Pd [26]) were observed. New nuclides 129Pm [27] and 139Tb [28] were studied using the X-γ coincidence technique with the helium-jet tape transport system. These studies are reviewed in Ref. [29]. Additionally, 69Kr [30, 31] was studied using a pulsed-beam technique in combination with a ΔE-E-Ereject particle telescope.

3.4 235Am

The new isotope 235Am [32] was produced by the reaction 238Pu(p,4n)235Am using 35 MeV protons from the proton linear accelerator at the Institute of High Energy Physics, CAS. A helium-jet multiple-target chamber system transported reaction products to a low-background area where they were collected on a tantalum disc. The collected products were dissolved in nitric acid, and americium isotopes were chemically separated from fission products and other actinides by a rapid procedure. From observed Np KX rays, 235Pu γ rays, and X-γ coincidence measurements, the new isotope 235Am was identified with a half-life of 15$±$5 min.

4 The Third Stage and Gas-Filled Recoil Separator SHANS Period

Beginning in 2010, all studies of heavy and super-heavy nuclei have been performed using the new gas-filled recoil separator SHANS (Spectrometer for Heavy Atoms and Nuclear Structure).

4.1 SHANS and Detection System

SHANS [33] is a gas-filled recoil separator with a QvDhQvQh configuration, where Q and D denote quadrupole and dipole magnets, and subscripts v and h refer to vertical and horizontal focusing directions, respectively. Its total length is 6.5 m with an angular acceptance of 25 msr. The dipole magnet has a maximum magnetic rigidity of 2.88 Tm and a bending angle of 52°. A silicon semiconductor detector box (Si-box) is installed at the focal plane of the separator. Figure 2 shows the layout of SHANS and the detection system at the focal plane.

Three 300-μm thick position-sensitive silicon detectors (PSSD) are mounted at the back of the Si-box as stop detectors for measuring implantation, fission fragments, and α particles. Eight non-position-sensitive silicon detectors are mounted in an open box arrangement around the strip detectors to detect escaping radioactive decay events. Three punch-through detectors are mounted behind the PSSD to provide veto signals for light particles passing through the strip detectors. A multi-wire proportional counter (MWPC) is mounted upstream of the Si-box as a timing detector. The time-of-flight (TOF) between the MWPC and PSSD distinguishes radioactive decay events in the PSSD from implantation events.

Event chains consisting of implanted evaporation residues (EVR) and their subsequent α decay and/or spontaneous fission are identified by the position-time correlation method. The data acquisition system implements digital signal processing using 16 14-bit flash Analog to Digital Converters (ADCs) from CAEN Costruzioni Apparecchiature Elettroniche Nucleari S.p.A. [34]. Preamplifier signal shapes from all detectors are independently recorded in traces tens of

microseconds long with 100-MHz sampling rate. The preamplifier rise time of about 40 ns enables resolution of fast decay signals following EVR implantation.

4.2 271Ds

The super-heavy nuclide 271Ds [35] was synthesized via the 208Pb(64Ni,n) reaction at a beam energy of 313.3 MeV and separated and identified by SHANS shortly after its commissioning in 2011. Using the separator coupled with a position-sensitive silicon strip detector, energy-position-time correlation measurements were performed for implanted nuclei and their subsequent decay α particles. One α-decay chain for 271Ds was established, with the α-particle energy and decay time measured to be 10.644 MeV and 96.8 ms, respectively, consistent with literature values [36, 37]. The nucleus 271Ds is the heaviest isotope synthesized and studied at IMP/CAS and in China to date.

4.3 Neutron-Deficient Actinides

With continuous development and improvement of SHANS and related devices, 10 new isotopes in the most neutron-deficient actinide region from Ac to Np have been synthesized for the first time and their decay properties studied.

Many mass models (e.g., macroscopic-microscopic models FRDM [38], Duflo-Zuker [39], WS4 [40], Hartree-Fock-Bogoliubov models HFB-24 [41, 42], UNEDF0 [43], and spherical relativistic continuum Hartree-Bogoliubov (RCHB) theory [44]) predict the existence of multiple neutron-deficient actinide isotopes not yet found experimentally. Our motivation for these studies is to search for new actinides, particularly U and Np isotopes around the proton drip line for the heaviest elements, and investigate their decay properties. Additionally, these isotopes lie in the region with the magic number N=126, making investigation of shell structure evolution and N=126 shell closure highly intriguing. These studies are challenging due to low production cross sections and short half-lives.

Table 1 lists experimental details of these studies, including reaction systems and corresponding production cross sections. Using fusion-evaporation reactions between beams of 36Ar, 40Ar and 40Ca and enriched isotopic targets of 169Tm, 176Hf, 180,182,183,184,186W and 185,187Re at various beam energies, nuclides from Ac to Np have been synthesized and their decay properties studied experimentally.

For Np isotopes, new nuclei 219,220,222-224Np have been synthesized with several prominent results. First, based on the measured α-decay energy of 219Np [57] and masses of 215Pa and 218U [62], a proton separation energy Sp=−301$±83*keV was determined, experimentally reaching the proton drip − line for Np. Thus, in the region of nuclei with Z* 83*, the proton drip line has been reached for all odd− Z isotopes up to Np. Second, applying digital signal processing techniques, the half− life of 222Np was determined to be only 380^{+260}__{110}$ ns [59]. As the time-of-flight of 222Np EVRs through SHANS is estimated to be ~1.2 μs, this sub-μs value may represent the shortest half-life measurable by SHANS. Third,

new experimental results have established α-decay systematics for Np isotopes around N=126, allowing the first test of N=126 shell closure robustness in Z=93 Np isotopes. By examining systematics of α-decay Qα values, half-lives, and reduced widths, the persistent rigidity of the N=126 magic number in Np isotopes is clearly recognized [58].

Figure 3 shows systematics of Qα values for ground-state to ground-state transitions in neutron-deficient $89 \leq Z \leq 93$ isotopes, and proton separation energies Sp for odd-Z Ac, Pa, and Np isotopes as a function of neutron number.

New uranium isotopes 214,215,216U [52–54] have been synthesized. Combining experimental data, improved α-decay reduced widths δ^2 for even-even Po–Pu nuclei near the magic neutron number N=126 have been deduced. To study proton-neutron interaction effects on α decay in this region, systematic trends were analyzed using the NpNn scheme. Strikingly, reduced widths of 214,216U are significantly enhanced by a factor of two compared to NpNn systematics for $84 \leq Z \leq 90$ and $N < 126$ even-even nuclei [52]. Figure 4 [Figure 4: see original paper] shows systematics of reduced widths for ground-state to ground-state α decays of even-even $84 \leq Z \leq 94$ isotopes as a function of neutron number and against NpNn for even-even Po to U isotopes. This abnormal enhancement is interpreted as resulting from strong monopole interaction between valence protons and neutrons occupying the $ $1f7/2 and 1f5/2 spin-orbit partner orbits, supported by large-scale shell model calculations.

5 Perspectives

In the near future, studies on heavy and super-heavy nuclei/elements will be performed at two facilities. Based on the existing gas-filled recoil separator SHANS, we will continue investigating neutron-deficient isotopes in the actinide and/or super-heavy nuclei region, including synthesizing new Pu and Am isotopes and studying their decay properties. Currently, a new facility named CAFE2 including SHANS2 is under construction, with synthesis of super-heavy elements and nuclei as our long-term goal.

CAFE2 (China Accelerator Facility for super-heavy Elements) is a new facility being constructed at IMP/CAS based on an upgraded superconducting linear accelerator. The new gas-filled recoil separator SHANS2 (Spectrometer for Heavy Atoms and Nuclear Structure-2) [63] at CAFE2 has a total flight path of 5.85 m with an ion-optical configuration of QvDQhQvD. The maximum magnetic rigidity is 2.5 Tm with maximum acceptable divergence angles of $±$70 mrad and $±$113 mrad in horizontal and vertical directions, respectively. Successful techniques for targets, differential pumping, recoil detection, and isotopic identification used on SHANS will also be applied to SHANS2.

Following commissioning of CAFE2 and SHANS2, efforts will focus on producing and synthesizing elements 119 and 120. Medium-mass beams such as 50Ti, 51V, 54Cr and 55Mn will be used to bombard radioactive actinide targets such as 243Am and 248Cm. As CAFE2 is a stand-alone facility devoted to this re-

search, sufficient beam time is expected, providing an opportunity to attempt measurement of new elements.

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