

GSI, Darmstadt, October 23, 2015 14th Workshop on



Recoil Separator for Superheavy Element Chemistry

supported by the Helmholtz Institute Mainz (HIM) and the JAEA ASRC's Reimei program

Final Program and Abstracts

October 22, 2015

٦	IME	TOPIC
1	9:00	Optional: Dinner at "Bürgermeister-Pohl-Haus", Wixhausen (registration required)

October 23, 2015 (Location: KBW Lecture Hall, GSI)

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09:00	Welcome	T. Stöhlker, Research director of GSI	
	Scientific highlights from gas-filled (and other) separators $igstarrow$	Chair: A. Türler (PSI/Bern)	
09:10	Status and perspectives of SHE syntheses at RIKEN GARIS	H. Haba (RIKEN)	2
09:35	First ionization potential measurement of heaviest actinides	T. Sato (JAEA, Tokai)	2
10:00	First observation of an atomic level for the element nobelium	M. Laatiaoui (HIM, Mainz)	3
10:25	Coffee		
10:45	Status on the chemical investigations of flerovium at TASCA	L. Lens (Univ. Mainz)	3
11:10	MARA, a new in-flight recoil separator for nuclear spectroscopy	J. Uusitalo (JYFL)	4
11:35	New short-lived 221 U and the mass-surface ear N = 126	J. Khuyagbaatar (HIM/GSI)	4
12:00	Enhanced fission stability of K isomers in superheavy nuclei: the case of ²⁵⁴ Rf	H. David (GSI)	5
12:25	Lunch		
13:15	Workshop Photo		
	Detector developments *	Chair: RD. Herzberg, Liverpool	
13:30	Diamond detectors, TI adsorption on quartz and E113	P. Steinegger (PSI)	5
13:50	Point-like contact Ge detectors for high resolution γ -spectroscopy	L. Harkness-Brennan (Univ. of Liverpool)	6
14:10	Opportunity with the new ALBEGA multi-coincidence detector	A. Di Nitto (Univ. Mainz)	6
14:30	Testing of the new SHIP focal plane detector system	A. Mistry (HIM, Mainz)	7
14:50	Coffee		
	(Old and) new avenues and technical developments for SHE st	Chair: M. Schädel, GSI	
	Realistic modeling for in-flight gas-filled recoil separators	J. Sarén (JYFL)	7
15:30	Radiochemical Investigation of the kinematics of multi-nucleon transfer reactions in $^{48}Ca + ^{248}Cm$ collisions at 1.1 x B	M. Götz (Univ. Mainz)	8
	Carbonyl chemistry developments for SHE studies	A. Türler (PSI)	9
16:10	Towards Cn and FI selenides: unexpected Cn-Se bond formation	N.M. Chiera (PSI)	10
16:30	Fundamentals of He future gas stopping cell for online measurements at TASCA	S. Götz (HIM, Mainz)	11
16:50	Development of a rapid solvent extraction apparatus for aqueous chemistry of the heaviest elements	Y. Komori (RIKEN)	12
17:10	Closing remarks		
17:15	End of TASCA 15 workshop		

• 15 minutes presentation plus 10 minutes discussion time

*15 minutes presentation plus 5 minutes discussion time



Present Status and Perspectives of SHE Syntheses at RIKEN GARIS

Hiromitsu Haba for the SHE synthesis collaboration at GARIS Nishina Center for Accelerator-Based Science, RIKEN, Wako, Saitama 351-0198, Japan

Present status and perspectives of superheavy element (SHE) syntheses with the GAs-filled Recoil Ion Separator (GARIS) at the RIKEN Linear Accelerator are reviewed.

So far, we have investigated production and decay properties of long-lived ²⁶¹Rf, ²⁶²Db, and ²⁶⁵Sg useful for chemistry studies with a gas-jet transport system coupled to GARIS [1–3]. In this workshop, we will present a new result on the production and decay properties of Bh isotopes in the ²⁴⁸Cm(²³Na,*xn*) reactions for future Bh chemistry after the GARIS separation.

Since an observation of the third decay chain of element 113 in the cold fusion reaction of ${}^{209}\text{Bi}({}^{70}\text{Zn},n){}^{278}\text{113}$ [4], we have started syntheses of the heavier SHEs by hot fusion reactions. The isotopes of element 116, ${}^{292}\text{Lv}$ and ${}^{293}\text{Lv}$ were produced in the ${}^{248}\text{Cm}({}^{48}\text{Ca},xn){}^{296-x}\text{Lv}$ reactions [5,6] with GARIS to confirm those observed at Dubna [7,8] and GSI [9]. In 2016, the ${}^{248}\text{Cm}({}^{50}\text{Ti},xn){}^{298-x}\text{118}$ reactions will be studied with a new gas-filled recoil ion separator, GARIS II [10].

References

- [1] H. Haba et al., Phys. Rev. C 83, 034602 (2011).
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First Ionization Potential Measurement of the Heaviest Actinides

T.K. Sato

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The first ionization potential (IP) is a fundamental physical and chemical property of an element. Information on the IP of the heaviest elements can provide better understanding of relativistic effects which are significantly noticeable for heavy elements. IP values of the heaviest elements above einsteinium (Es, Z = 99), however, have not been measured so far due to their low production rates and short half-lives. We have developed a novel method for IP measurement based on a surface ionization process [1].

In my talk, I will report on the first experimental determination of the IP values of the heaviest actinide elements, No, Md and Fm..

References

[1] T.K. Sato, et al. Nature 520, 209-211 (2015).



First observation of an atomic level in the element nobelium

M. Laatiaoui Helmholtz-Institute Mainz (HIM), Mainz, Germany

Laser spectroscopy of elements beyond fermium (Z = 100) is nowadays considered to be one of the most fascinating and simultaneously challenging tasks in atomic physics. In particular, the online production rates of a few atoms per second at most render any optical spectroscopy in that region of the chart of nuclei extremely difficult. However, such studies are of special importance as they would shed light on the impact of relativisticand QEDeffects on the electronic structure of such atoms, for which so far no experimental data exist. Laser spectroscopy of the element Nobelium (Z = 102) has long been anticipated at GSI in Darmstadt [1]. In my talk I will summarize the pioneering work in this field of research, and report on the first observation of an atomic level in ²⁵⁴No.

References

[1] H. Backe et al., EPJD 45, 99 (2007).

Status on the chemical investigations of flerovium at TASCA

Lotte Lens¹

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The experimental determination of chemical properties of element 114 (flerovium, Fl) is among the hottest topics in current transactinide element research. The question whether Fl behaves like a noble gas or like a noble metal could not be answered unambiguously to date, due to limited statistical results [1, 2].

This talk covers advanced experiments, conducted by GSI Darmstadt, studying the chemical properties of Fl, using the gas-filled separator TASCA for preseparation.

References

[1] R. Eichler, et al. Radiochim. Acta 98, 133-139 (2010).

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MARA, Mass Analyzing Recoil Apparatus, a new tool at JYFLACCLAB

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A new in-flight mass separator has been constructed at Jyväskylä Accelerator Laboratory (JYFLACCLAB). The configuration of this separator is QQQEDMD [1] which is different from the existing symmetric devices [2, 3, 4] having two electric dipoles (ED) and one magnetic dipole (MD) in between. Also in symmetric devices quadrupole doublets (QQ) are used at the beginning (and at the end) of the separator. With the configuration used at MARA the separator could be made shorter (space limitations) and the cost of the separator is reduced. MARA will be used for studies performed at the proton drip line below mass number 140 using symmetric fusion evaporation reactions (or slightly in inverse kinematics). It will be a complementary device to the existing gas-filled recoil separator RITU [5] which is well suited for studies of heavier elements using asymmetric reactions. Construction of the MARA device has now been finished. The ion-optics of the separator has been studied using different alpha sources. First beams has been taken to the separator and fusion evaporation recoils have been successfully transported to the focal plane of MARA.

In this work the MARA design will be presented. The status report of the separator as well as the future plans will be given.

References

[1] J. Saren, Research Report No. 7/2011, Department of Physics, University of Jyväskylä.

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- [3] H. Ikezoe *et. al.*, NIM A **376** 420 (1996).
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The new short-lived isotope ²²¹U and mass-surface near N =126

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The shell structure of the atomic nucleus is one of the fundamental pillars of nature. To date many features of the nuclear structure are established, especially in the nuclei around the proton and neutron numbers Z, N=2, 8, 20, 28, 50, 82 and N =126 where an enhanced stability against any type of ground-state radioactive decay occur. The evolution of the N=126 shell closure towards higher Z above Pa (Z =91) is yet poorly examined. An absence of data on U isotopes with N = 128-130 prevents performing comparative analyses of the empirical observables for the presence of the shell closure, as they were done in lighter elements.

I will show the results of a TASCA experiment where two of those isotopes, 221U (N =129) and 222U (N=130), were produced as evaporation residues of the fusion reaction 50Ti+176Yb at the gas-filled recoil separator TASCA. Synthesis and detection of these unstable heavy nuclei and their descendants were achieved thanks to a fast data read-out system.

The evolution of the N = 126 shell closure towards higher Z and its influence on the stability of uranium isotopes made within the framework of alpha-decay reduced width will be presented.



Enhanced fission stability of K isomers in superheavy nuclei: the case of ²⁵⁴Rf

Helena May David Argonne National Laboratory*

The question of the maximum number of nucleons that can reside in a nucleus has driven a major effort in both the production and study of nuclei at the limits of mass and in their theoretical description. Contemporary models disagree on the location of the long-predicted island of stability for spherical superheavy nuclei, which remains beyond current experimental reach. Transfermium nuclei close to the Z=100, N=152 deformed shell gaps are, however, accessible experimentally, and information on their structure can provide stringent tests for existing models. In particular, the location of high-K isomers can be used to deduce single-particle energies. Twoquasiparticle (2qp) K isomers have been observed in several even-even N=150 isotones, from ²⁴⁴Pu to ²⁵²No. I will focus on the striking case of the very fissile nucleus ²⁵⁴Rf - the heaviest known N=150 isotone, in which a 2qp and 4qp isomer have been recently observed using the Argonne Fragment Mass Analyzer and the Berkeley Gas-filled Separator. Surprisingly, the decay of the 2qp isomer is four orders of magnitude faster than found for analogous isomers in lighter N=150 isotones. The 4qp isomer is longer lived than the ²⁵⁴Rf ground state, which decays exclusively by spontaneous fission. Also unexpectedly, no evidence was found for a fission branch from either isomer, resulting in unprecendented fission hindrance relative to the groundstate fission.

*Present address: GSI Darmstadt

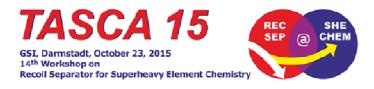
Diamond detectors, TI adsorption on Quartz and E113

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The surface interaction of thallium on quartz was measured in an on-line isothermal vacuum chromatography experiment at the on-atom-at-a-time level, yielding an adsorption enthalpy of $-\Delta H_{ads}(Tl \text{ on } SiO_2) = 158 \pm 3 \text{ kJ} \cdot \text{mol}^{-1}$ [1]. This differs significantly from early measurements of the same physicochemical system [2] and from the adsorption enthalpy for TIOH on quartz [3]. Here we present the experimental approach and briefly discuss the outcome in the light of the mentioned discrepancies. The successful vacuum chromatography experiment in combination with a first-ever on-line α -spectroscopy detection stage using diamond detectors, allow for a change of focus towards a superheavy element chemistry experiment with E113.

References

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- [2] B. Eichler, ZfK-Report 346 (1977).
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Point-like contact germanium detectors for high-resolution gamma spectroscopy

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The excellent energy resolution of point-like contact germanium detectors offers the potential for exciting new measurements in gamma-ray spectroscopy. Such detectors are already in use in the ALBEGA system for alphabeta-photon multi-coincidence spectroscopy of chemically separated samples. This talk will outline the principles of point contact detectors and introduce the types of sensors available, ranging from existing commercial products to the next-generation SIGMA detector.

Opportunity for next generation experiments with the new ALBEGA multi-coincidence detection setup

A. Di Nitto Johannes Gutenberg-University, 55099 Mainz, Germany

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The present knowledge of the nuclear structure of superheavy nuclei (SHN) is still scarce, in spite of large efforts devoted to in the last decades [1]. This is mainly due to the low production rate [2]. The most successful method for producing the SHN nuclei is to use heavy-ion induced fusion-evaporation reactions performed with in-flight recoil separators in combination with a detection setup. An improvement of the background conditions was observed applying a chemical isolation system after separator [3]. In order to collect a more complete spectroscopic dataset in such experiments, a next generation detection system ALBEGA (for measurements of ALpha-BEta-GAmma decays after chemical isolation) was recently built at GSI. ALBEGA is dedicated to simultaneous measurements of α -particles, electrons, photons and fission fragments. The setup, which comprises of a gas channel, Si detectors and Ge detectors, has recently been built and tested. First experimental data in comparison with GEANT4 simulations, and an outlook on future applications will be presented.

References

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Testing of the new SHIP focal plane detector system

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Study of the structure of heavy and superheavy nuclei requires continuous advancement of specialized spectroscopic detection systems and analysis techniques. A new focal plane detection system has been designed and developed at GSI for the SHIP separator [1]. It consists of a DSSD implantation detector surrounded by 4 SSSD detectors on each side in a box formation with data acquisition using the flash ADC module FEBEX for digital signal processing. Currently, testing is underway online at SHIP. In my talk I will present preliminary measurements from heavy actinide nuclei demonstrating the performance of the device.

References

[1] D. Ackermann, GSI Annual report (2015).

Realistic modeling for in-flight gas-filled recoil separators

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A new computer code library [1] has been developed for modeling of a gas-filled recoil separator. The code implements a full Monte-Carlo approach for particle tracking through a separator. The magnetic fields are interpolated from 2D/3D field maps or calculated from an ideal sharp edge model. All the elements can be rotated and translated freely in 3D space. The code takes into account the charge-exchange collisions, scattering of the recoils, energy losses and effects arising from the mean free time between the subsequent collisions. These processes are discussed and a few adequate models are reviewed. Results of the modeling of the ¹⁵⁰Sm(⁴⁰Ar,4n)¹⁸⁶Hg reaction are compared to the experimental results measured with the RITU separator [2]. The comparison shows a remarkably good agreement after tuning of the few model parameters. The code is intended to be publicly available in the near future.

References

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Radiochemical investigation of the kinematics of multi-nucleon transfer reactions in ${}^{48}Ca + {}^{248}Cm$ collisions at 1.1 x B

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The renewed interest in multi-nucleon transfer reactions as a promising tool for the production of neutron-rich transactinide isotopes [1,2,3,4] has motivated us to perform a ⁴⁸Ca+²⁴⁸Cm bombardment at an incident energy 10% above the Coulomb barrier to study angular distributions and recoil ranges of Cf through Fm isotopes. These were stopped in Ni catcher foils covering slices of laboratory angles between 17° and 65° and differential recoil ranges between 3 μ m and 6 μ m. The foils were dissolved in dilute nitric acid, tracer activities of Lu and Eu were added for chemical yield determination, and the activities were co-precipitated with Fe(OH)₃ using ammonia to remove Ni, separated from Fe, Pa, U, Np, and Pu on an anion-exchange column in 8 M HCl, separated from Th and Ra on a cation-exchange column in 2.25 M HCl, and separated on a cation-exchange column with 0.12 M α -HiB and 0.14 M α -HiB at pH=4.80 into a Fm(Es) fraction and a Cf(Bk) fraction, respectively. These were prepared for α -particle spectroscopy and spontaneous-fission counting. The investigated parameters are the centroids of the post-neutron emission isotope distributions and their displacement from the most probable primary fragment mass numbers resulting from Volkov's generalized Q_{gg} systematics including corrections for the breaking of nucleon pairs in the multi-nucleon transfer process [3,5], the angular distributions, and the total kinetic energy loss TKEL. The results for isotopes of transcurium elements up to Fm are being evaluated and will be presented at the conference.

References

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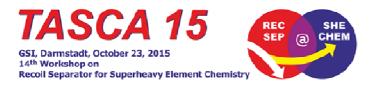
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Holland, Amsterdam) Vol. II, 363 (1974).



Carbonyl chemistry developments for SHE studies

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+ recently at TRIUMF, Vancouver, BC V6T 2A3, Canada

We intend to summarize here the preparation status for experiments envisaging to assess for the first time directly the bond dissociation enthalpy in a transactinide molecule, Sg(CO)₆. On-line decomposition experiments with ¹⁰⁴Mo and ¹⁶⁴W performed at Bern University and RIKEN GARIS revealed a decomposition behavior of the corresponding hexacarbonyl compounds on silver surfaces correlated to their first-bond dissociation enthalpy (see figures) [1]. The 23 kJ/mol bond stability increase between Mo(CO)₆ and W(CO)₆ [2] lead to a decomposition temperature shift of about 100 K. A superimposed chromatographic adsorption-decomposition model combined with theoretically predicted FBDE for Sg(CO)₆ [3], allowed to predict an expected decomposition behavior of Sg(CO)₆ in an experiment similar to the model experiments, and thus, the design of a corresponding transactinide experiment.

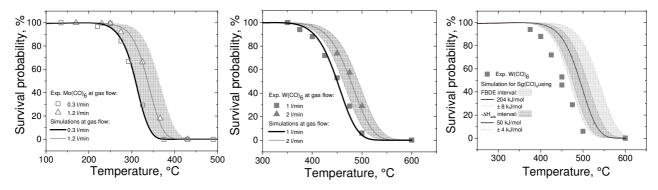


Fig. 1. Experimental decomposition curves of Mo(CO)₆ and W(CO)₆ at different gas flow rates (symbols) together with simulated decomposition curves (lines) using literature data for ΔH_{ads} and FBDE. The kinetic preexponential factor was deduced for all hexacarbonyl decompositions as $A = 8.6 \cdot 10^5$. The shaded area shows the e.g. response of the model to a variation of ΔH_{ads} within ±4 kJ/mol [4]. The predicted behavior of Sg(CO)₆ in the same experiment including the uncertainties in FBDE [3] and ΔH_{ads} [4].

References

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Towards selenides of the SHE Copernicium and Flerovium: Unexpected Cn-Se bond formation

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Extrapolative predictions revealed that for the selenides formation the stability trends in group 12 and 14 are predicted to be opposite: The formation of FISe is expected to be favored, while the interaction between Cn and selenium is expected to be a weak physisorption [1]. The difference in these trends indicates the selenium surface to be a good candidate for comparative studies of the chemical behaviour of copernicium and flerovium. In preparation of the chemical investigations of these superheavy elements, two different selenium allotropes red amorphous selenium and trigonal selenium- were used as stationary surfaces in isothermal model experiments with ¹⁹⁷Hg. The performed Monte-Carlo simulations were in good agreement with the experimental results, assuming as lower interaction limit $-\Delta H_{ads}^{Hg}(a-Se) > 80$ kJ/mol for a 95% red amorphous Se surface coverage, and $-\Delta H_{ads}^{Hg}(t-Se) < 45$ kJ/mol for a 50% trigonal Se and 5 % red amorphous Se surface coverage. The results indicate a severe kinetic hindrance of the Hg / trigonal Se reaction taking into account the thermodynamic stability of HgSe [2]. To prevent the transformation of the metastable red amorphous selenium to the most stable trigonal crystal structure, a vapor transport deposition (VTD) technique for homogeneous coating of quartz tubes and silicon detector surfaces was optimized [3]. A device for the storage of columns and diodes covered with red amorphous Se was developed as well. First on-line test experiments using the COLD detector array with Se covered detector surfaces were performed. The Se surface crystallization, monitored by the ¹⁸⁵Hg deposition pattern, revealed an advanced crystallization of the thin red a-Se surface on the detectors after 3 weeks of storage. Two events detected on Se-covered detectors were attributed to ²⁸³Cn. Hence, first indication that Cn reacted with trigonal Se with a probability >95% was found. Monte Carlo simulations revealed a $-\Delta H_{ads}^{Cn}(t-Se) > 48$ kJ/mol. Despite the CnSe formation is expected to be thermodynamically less favored, these first results reveal a lower kinetic formation hindrance compared to the interaction of Hg with t-Se. Further on-line studies with Hg, Cn and Fl on both trigonal and amorphous selenium surfaces are envisaged in 2016.

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Fundamentals of the future gas stopping cell for online measurements behind TASCA

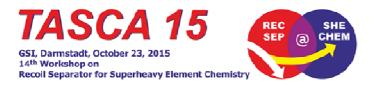
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The interest to produce heavier elements is closely linked to answer the question "Which is the heaviest element than can exist?". This quest is the force behind search experiments like element 114 at the GSI Helmholtzzentrum für Schwerionenforschung in Darmstadt, Germany. One of the long-term objectives of the GSI is the enabling of chemical studies of elements beyond flerovium (Fl). Here already established technologies and facilities available at GSI appear ideal. The production and separation of the elements of interest can be performed with the well-studied preseparator TASCA. With the current setup of TASCA and COMPACT the most time-consuming step is flushing the gas stoppers, also known as recoil transfer chamber (RTC). RTC-based setups have a typical extraction time of hundreds of ms. To get access to shorter-lived isotopes and thus eventually to elements 115 and beyond, a faster technique is needed. The RTC is the rate limiting factor in the current configuration and therefore the logical consequence is to modify or replace this. At the moment two different designs of gas stoppers be can be installed at particle accelerator facilities. In the first design a gas flow is used to extract the ions from the relatively simple gas-filled chamber (RTC) [1-3]. The extraction efficiency of these design is high, but unfortunately the resulting beam of thermalized products has a has a very large and poorly characterized emittance. The second design uses radiofrequency (RF) oscillating electric fields to extract the ions [4-8]. As a result, short extraction times and high extraction efficiencies are achieved, but the used RF systems are extremely complex.

A new gas stopper has been developed, which uses a DC-field gradient (typical 10 V/cm) to guide the thermalized ions in approx. 50 mbar gas volume to the detector-exit. The funnel focuses the ions toward the exit hole with a diameter of 0.6 mm. Such a system should be characterized comprehensively in accordance with the common methods and strategies. This involves the characterization of the extraction times and extraction efficiencies and the influence of operating parameters on the extraction process. The development of appropriate interfaces for connecting such a gas stopper between TASCA and COMPACT is a prerequisite for its practical use. The suitability as thermalization unit for TASCA must be investigated and the appropriate analytical figures of merit are to be determined.

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Development of a rapid solvent extraction apparatus for aqueous chemistry of the heaviest elements

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Aqueous chemistry studies of superheavy elements (SHEs) have been so far carried out up to element 106, Sg. Following the pioneering studies for Sg in 1990s [1], however, there have been no reports on the aqueous chemistry of Sg and heavier SHEs. It is extremely difficult to perform aqueous chemistry experiments with SHE nuclides due to their decreasing production yields and half-lives with increasing the atomic number. Further, a huge amount of background radioactivities of by-products become unavoidable in detection of α particles and SF fragments from the SHE nuclei of interest. To overcome these problems, we plan to use the RIKEN GAs-filled Recoil Ion Separator (GARIS) as a preseparator for the chemistry experiments of SHEs [2].

Toward the aqueous chemistry of Sg and element 107, Bh, we have been developing a rapid solvent extraction apparatus coupled to the GARIS gas-jet system [2] (Fig. 1). This new chemistry apparatus consists of a continuous dissolution apparatus (Membrane DeGasser: MDG) [3], a Flow Solvent Extractor (FSE), and a flow liquid scintillation detector for α /SF-spectrometry. Recently, we investigated performances of MDG and FSE using Tc and Re isotopes produced in the ^{nat}Mo(*d*,*xn*) and ^{nat}W(*d*,*xn*) reactions, respectively, at the RIKEN AVF cyclotron. In the workshop, current status and perspectives of the SHE aqueous chemistry project at GARIS are reviewed.

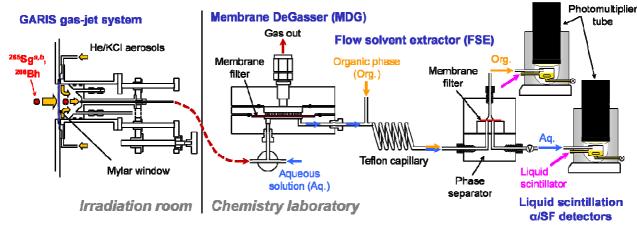


Fig. 1. Layout of a rapid solvent extraction apparatus coupled to the GARIS gas-jet system.

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