FACILITY UPGRADE FOR SHE RESEARCH AT RIKEN NISHINA CENTER*

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Received 27 November 2022, accepted 16 January 2023, published online 22 March 2023

In 2016, the RIKEN Nishina Center (RNC) commenced a new comprehensive superheavy element (SHE) research program, the "SHE project". The project aimed at synthesizing a new superheavy element, $Z = 119$, through a hot fusion reaction $^{51}V+^{248}Cm$ constructing a superconducting RIKEN linear accelerator (SRILAC) and a new superconducting electron-cyclotron-resonance ion source (SC-ECRIS) to boost the final energy and intensity. A gas-filled recoil ion separator (GARIS-III) suitable for detecting the residues of the hot-fusion reaction was also constructed. The SHE project and its commissioning results are briefly described.

DOI:10.5506/APhysPolBSupp.16.4-A10

1. Prologue

Before introducing the SHE project, one of the authors, Sakai, would like to pay his respects to the late Prof. Sigurd Hofmann by mentioning some personal memories of him and Sakai. Although we have known each other for a long time, in 2017, when we started working together as members of the Joint Working Group (JWG) organized under the International Union on Pure and Applied Physics (IUPAP) and International Pure and Applied Chemistry (IUPAC), we became deeply acquainted.

The criteria established by the Transfermium Working Group (TWG) in 1993 [1] were used to certify the discovery of superheavy elements discovered since then. Nine superheavy element candidates for $Z = 110$–118 were experimentally observed and reported between 1994 and 2004. The Joint Working Party (JWP) formed under IUPAP and IUPAC reviewed these discoveries based on the criteria. It was in 2016 that JWP approved the claim of the discovery of $Z = 113$, 115, 117, and 118, which remained till

* Presented at the Zakopane Conference on Nuclear Physics, *Extremes of the Nuclear Landscape*, Zakopane, Poland, 28 August–4 September, 2022.

the last [2, 3]. During these reviews, it was recognized that new criteria might be needed to review discovery claims for $Z > 118$, which utilize newly developed experimental techniques and equipment. Concerned about this situation, IUPAP and IUPAC thus, organized JWG in 2016 to establish new criteria and appointed Sigurd as the Chair. Subsequently, its kick-off meeting was held in Egelsbach, Germany, in 2017. Figure 1 shows the memorial photo of the meeting. Sigurd put his outstanding knowledge and abilities into drafting new criteria. Sakai had much fun working with him and appreciated learning all kinds of superheavy element physics!

Fig. 1. JWG kick-off meeting held at Egelsbach, Germany on May 21, 2017. From left to right: Sigurd Hofmann (GSI) Chair, Sergey N. Dmitriev (JINR), Jacklyn M. Gates (LBNL), Natalia Tarasova (IUPAC President), Bruce McKellar (IUPAP President), James B. Roberto (ORNL), Hideyuki Sakai (RIKEN) Vice-Chair, and Claes Fahlander (Lund University).

Sigurd's efforts resulted in the first version of the criteria released to the public in 2018. After a year of public review, the final version was finally published with a few minor corrections in 2020 [4].

Although Sigurd is well known as the leading scientist in discovering three elements, Ds, Rg, and Cn, the JWG report of criteria 'On the discovery of new elements' [4] would also stay as the legacy of the field.

2. Introduction

RIKEN has a long nuclear-science research tradition pioneered by Yoshio Nishina [5] who constructed the first cyclotron (26-inch) in 1937 outside the United States (second in the world). The discovery of the element nihonium [6] was one of the recent epoch-making achievements of the RNC. It was synthesized through the cold fusion reaction $^{209}\mathrm{Bi}(^{70}\mathrm{Zn}, n)^{278}\mathrm{Nh}$. After the discovery of nihonium, RNC set up the next research target as the

"SHE project", aiming to synthesize element $Z = 119$ through the hot fusion reaction of ^{248}Cm(^{51}V$, xn)^{299-x}$119 with $x = 3$ or 4. Table 1 lists the recent theoretical calculations of the evaporation residue (ER) cross-section, σ_{ER}, of the ^{248}Cm(^{51}V$, xn)^{299-x}$119 reaction for the $x = 3$ and 4 channels. Although the theoretical prediction strongly depends on the assumed theoretical models, σ_{ER} can be of the order of 10 fb or less. The approximate bombarding energy required for a ^{51}V beam to synthesize $Z = 119$ via the ^{51}V + ^{248}Cm reaction can be estimated through a simple Coulomb barrier height calculation assuming appropriate radii for ^{51}V and ^{248}Cm or by utilizing the systematic trend of the barrier height distribution measurements on ^{248}Cm by Tanaka *et al.* [7, 8]. Both estimates indicate that the required beam energy of ^{51}V is approximately 6 MeV$/u$ ($\sim$300 MeV) in the lab system. Note that the present heavy-ion linac (called RILAC) can provide a beam of up to 5.5 MeV$/u$.

Table 1. Theoretical calculations of the evaporation residue.

^{248}Cm(^{51}V$, xn)^{299-x}$119 channel x	Cross section [fb] 3n	4n	Ref.
Ghahramany (2016)	20	100	[9]
Zhu (2016)	6	11	[10]
Adamian (2018)		12	[11]
Manjunatha (2019)	4		[12]*
Siwek-Wilczyńska (2019)	3	6	[13]
Aritomo (2020)	20 at $E^* = 20$ MeV		[14]
Lv (2021)	9.8	1.3	[15]

*The value is read from Fig. 16. There are typos in this paper. Thus the value should be treated with caution.

The SHE project is thus the upgrade of the RILAC accelerator by replacing in part a superconducting RIKEN linear accelerator (SRILAC) to increase the final beam energy from 5.5 MeV$/u$ to 6.5 MeV$/u$ and build a new superconducting electron-cyclotron-resonance ion source (SC-ECRIS) operating at a higher RF frequency to increase the beam current, see Table 2 Construction of GARIS-III dedicated to the hot-fusion reaction was also planned.

The present contribution describes the SHE project taken mainly from Ref. [16].

Table 2. Goal specifications: before and after upgrade.

Upgrade name	Before RILAC	After SRILAC
Number of tanks	12DTLs	8DTLs, 10SC–QWRs
Frequency [MHz]	37.75/75.5	36.5*/73.0
Total Acc. V [MV]	25 ($A/q = 5$)	39 ($A/q = 6$)
Beam current [$p\mu$A]	0.5	> 2.5

*36.5 MHz is the fundamental frequency of the RF system of the RIBF accelerators.

3. Energy and intensity upgrade of RILAC

For the synthesis of $Z = 119$, a ^{51}V beam with more than 6 MeV/u is required, necessitating an energy upgrade. A superconducting-linac (SC-linac) was proposed [17] for increasing the beam energy from 3.6 MeV/u to $\sim$ 6.5 MeV/u. Given the limited space in the existing RILAC building, SRILAC was installed, replacing four drift-tube-linacs (DTLs) out of the six DTLs marked as CSM in the upper diagram in Fig. 2.

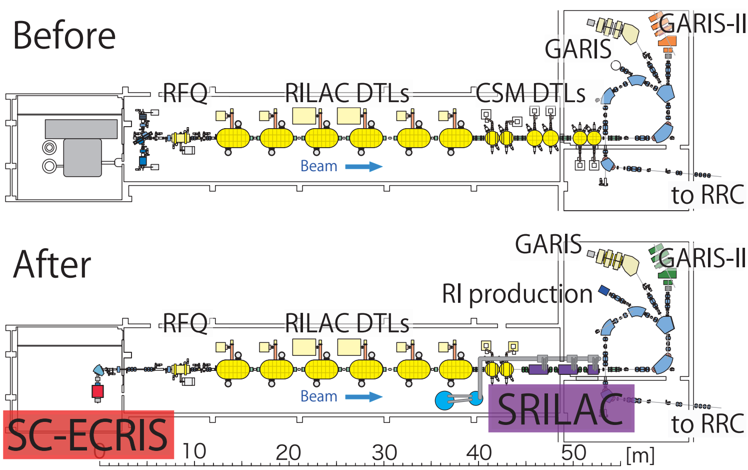

Fig. 2. Overview of RILAC and SRILAC.

SRILAC comprises three cryomodules (CM1, CM2, and CM3). The schematic drawing of CM1 (CM2) is shown in Fig. 3. CM1 and CM2 contain four SC-quarter-wavelength resonators (SC-QWRs) each, while CM3 is composed of two SC-QWRs only. The SC-QWR (SC-cavity) shown in Fig. 4 [18] is made from highly purified Nb sheets. The cavity performance was validated by measuring the quality factor (Q_0) related to the power dissipation. The typical Q_0 value of the ten bulk cavities measured at 4.2 K is $\sim 1 \times 10^9$ at a frequency (f) of 73 MHz. A stable $^{51}V^{13+}$ beam with energy ranging from 4.2–6.3 MeV/u was successfully accelerated and delivered.

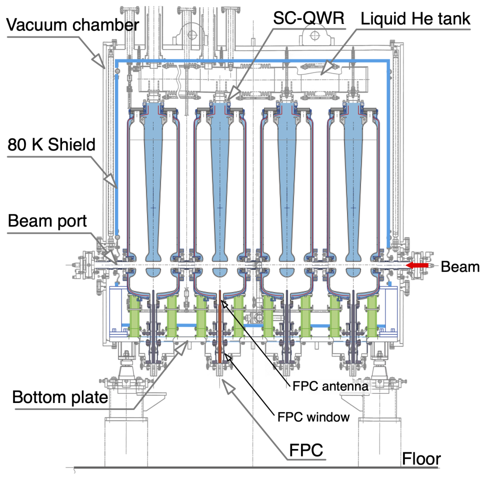

Fig. 3. Side-view of the cryomodule.

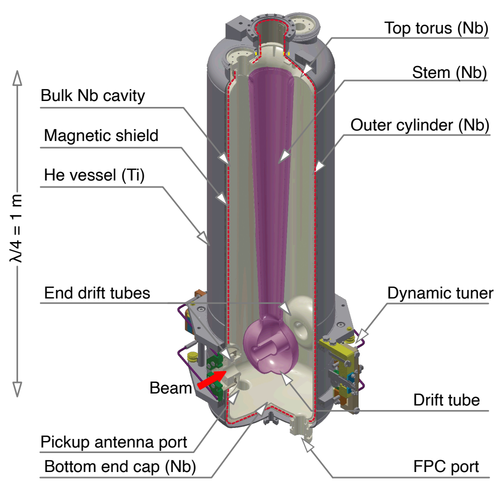

Fig. 4. Schematic view of the SC-QWR for SRILAC.

As an intensity upgrade, a new SC-ECRIS was constructed [19] to increase the beam current by at least five times than that used in the Nh synthesis experiment. The ion source for SRILAC is a duplication of the RIKEN 28-GHz SC-ECRIS [20] developed for producing an intense uranium beam together with a new injector linac (RILAC2) [21] for the RIBF. To provide a metal ion such as vanadium for a period of one month, an ion-source structure with a pair of high-temperature ovens was developed and installed [22]. An intense $^{51}V^{13+}$ beam is being produced from the ion-source for a period of nearly a month, typically $\sim$ 160 $e\mu$A ($\sim$ 12 $p\mu$A), which exceeds the targeted beam current for the experiment.

4. GARIS-III

A new gas-filled recoil ion separator GARIS-III suitable for hot-fusion reaction products was designed and constructed. The magnet configuration for GARIS-III includes two dipole magnets (D1 and D2) and three quadrupole magnets (Q1, Q2, and Q3) as shown in Fig. 5.

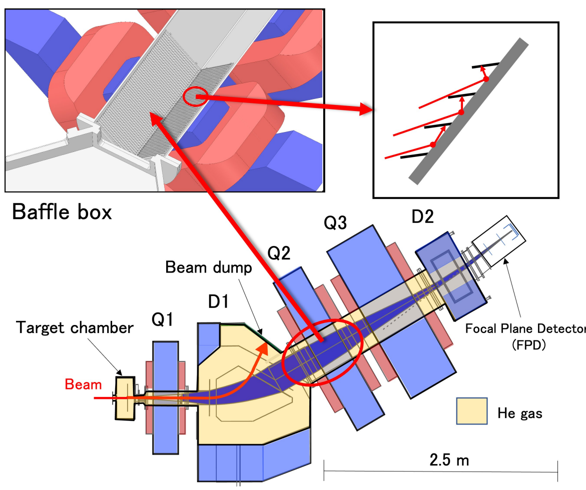

Fig. 5. (Color online) Plan view of GARIS-III. Top left inset: the baffle fin, magnet coil, and iron yoke are indicated in dark gray, reddish-brown, and blue, respectively.

A new aspect of GARIS-III compared to the GARIS-II [23] is a baffle box [24]. Inset on the top left is the 3D cross-sectional drawing of the baffle box within the vacuum chamber at the Q2 position. Inset on the top right is the cross section of the baffle with certain background particle trajectories as an example.

Figure 6 shows a layout of the target area consisting of the differential pumping section (DPS), N_2-gas-jet curtain [25], and target chamber. In the DPS, seven turbomolecular pumps (TMP) with an evacuation speed of 350 l/s and a mechanical booster pump (MBP) with an exhaust rate of 280 l/s are employed. A device called 'N_2-gas-jet curtain' was newly installed between the differential pumping section and the target chamber (filled with

30–100 Pa of He gas) to reduce the He-gas flow from the target chamber to the differential pumping section. The N_2-gas-jet curtain is highly effective in achieving better vacuum at the superconductive cavity section.

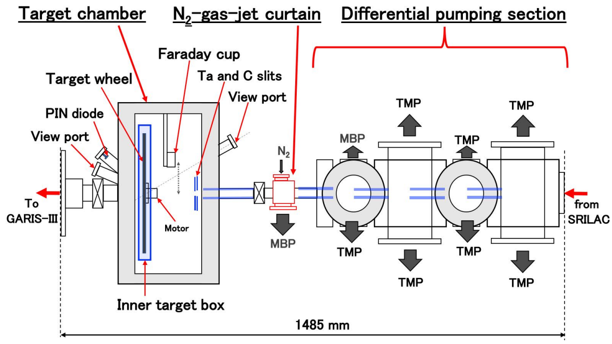

Fig. 6. Layout of the target area.

The validity of the whole system related with the GARIS-III experiment was partly confirmed by measuring the excitation functions of the ^{208}Pb(^{40}Ar$, xn)^{248-x}$Fm reactions [26]. The results for the $2n$ to $4n$ evaporation channels are plotted in Fig. 7 along with those measured at GSI [27]. As both results are highly consistent, it can be concluded that the fundamental properties of GARIS-III are confirmed [26]. The results were compared with the theoretical predictions by the nuclear reactions video (NRV) code [28] and the statistical fusion evaporation code, HIVAP [29]. It is interesting to note that the calculations reproduce the experimental results well.

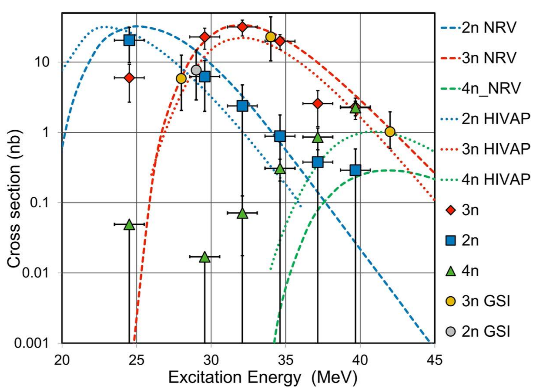

Fig. 7. Excitation functions of the ^{208}Pb(^{40}Ar$, xn)^{248-x}$Fm reactions. The results are from Ref. [26]. The reference data for the $2n$- and $3n$-channels are the GSI results [27]. The calculations are based on NRV and HIVAP. See the text for details.

5. Target preparation

Preparation of the ^{248}Cm target in the form of $^{248}Cm_2O_3$ plays an essential role in synthesizing $Z = 119$. The $^{248}Cm_2O_3$ target with the backing foil needs to withstand the exceedingly severe heat produced by the ^{51}V beam energy loss. Although the severity depends on the experimental conditions, the heat power would, for example, amount to 10 W for a 1 $p\mu$A beam with an energy loss of 10 MeV. Due to such high power, even under high-speed rotation, the temperature of the target/backing foil increases rapidly to 500–1,000°C depending on the surrounding cooling state, rapidly damaging the target/backing foil. Thus, it is necessary and inevitable to develop a target and a backing material that withstands the intense beam for an extended period, for the new SHE experiments.

Large and uniform $^{248}Cm_2O_3$ targets are routinely fabricated through a molecular plating method [30]. This large target wheel is located in a water-cooled target box in GARIS-III and rotates using a motor at 2,000 rpm in He atmosphere (33–73 Pa) during irradiation. Figure 8 shows the 30 cm diameter target wheel with sixteen sector targets.

Various backing materials such as Be, C, Ti, or Mo with several different thicknesses are being explored under actual experimental conditions.

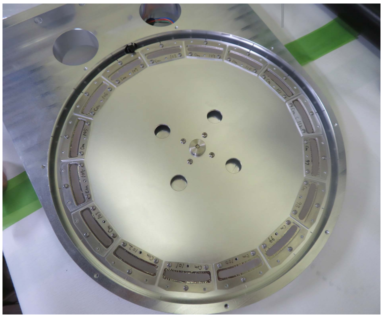

Fig. 8. Photograph of the rotating wheel with sixteen ^{248}Cm sector targets in the semiclosed inner-target box. The cover plate of the inner-target box is removed to display the interior.

6. Search for $Z = 119$

6.1. Selection of the optimal bombarding energy

One of the most decisive quantities that influence the yield of a new element through the hot-fusion reaction is the optimal bombarding energy ($E_{\rm opt}$). $E_{\rm opt}$ is dominated by the Coulomb barrier height energy B_0, which can be experimentally deduced from the excitation function of the quasielastic (QE) scattering cross section $d\sigma_{\rm QE}$, comparing to the Rutherford scattering cross section $d\sigma_{\rm Ruth}$. The ratio $d\sigma_{\rm QE}/d\sigma_{\rm Ruth}$ is denoted as $R(E)$.

At low incident energy, the incident flux is almost entirely reflected by the Coulomb barrier, resulting in $R \sim 1$ ($d\sigma_{QE} \sim d\sigma_{Ruth}$). As the incident energy increases, the incident flux is absorbed by various channels, such as deep-inelastic scattering or fusion, resulting in $R \to 0$. Thus, the $R(E)$ sensitively probes the Coulomb barrier height energy. The B_0 value is conveniently defined by $R(E_{cm} = B_0) = 0.5$ [7]. Note that the $d\sigma_{QE}$ measurement at $\theta_{lab} \sim 180°$ is significant because the reaction is dominated by the angular momentum $\ell \sim 0\hbar$, which is a critical component leading the system to fuse.

Tanaka *et al.* [31] measured the QE backscattering cross sections at $\theta_{lab} \sim 180°$ for the $^{51}V + ^{248}Cm$ system with GARIS-III when SRILAC became operational and obtained the $R(E)$ distribution (see Fig. 5 in Ref. [31]). The deduced B_0 value was 225.6 ± 0.2 MeV. The final ^{51}V beam energy (E_{opt}) can be determined considering the B_0 value, the side collision effects due to the shape deformation [7, 31], and the loss of the beam energy in the backing material and $^{248}Cm_2O_3$ target material.

6.2. Element-119 search with GARIS-III

The search for the $Z = 119$ element through the $^{51}V + ^{248}Cm$ hot-fusion reaction commenced in 2020 with GARIS-III after the completion of SRILAC.

For full-scale measurement, a considerable quantity of target material ($^{248}Cm_2O_3$) is needed to fill the target wheel as shown in Fig. 8. The required quantity of highly enriched $^{248}Cm_2O_3$ was provided to the RNC under the material transfer agreement between the RNC and ORNL.

The isotopes of new element $^{296}119$ and/or $^{295}119$ can be identified as the evaporation residue implanted in a pixel of the double-sided silicon strip detector (DSSD) by observing seven sequential α decays in a chain (seven generations) under an ideal situation. However, such identification becomes problematic when the background particles accidentally enter the same pixel and mimic the expected α-decay energy while waiting for a cascading α decay. The background α-particle-like accidental events were estimated to be 6.9×10^{-4}/s at a beam intensity of 2 $p\mu$A for an energy range of 8–15 MeV produced in a 2×4 mm^2 pixel of DSSD in a focal plane detector (FPD) [32], based on the number of events observed in the pixel but not observed in ToF detectors. This low-accidental event rate enables the identification of the synthesized element 119 with sufficient certainty under the current experimental conditions.

Special attention has been paid to the target-beam spot shape. The shape of the ^{51}V beam on the rotating target is carefully adjusted to ensure a uniform distribution of typically 8 mm $\times$ 1–3 mm in the horizontal and vertical directions, respectively. Figure 9 is a snapshot of the target area from the downstream viewing port of the GARIS-III target chamber. See Fig. 6 for the position of the downstream viewing port. A high-intensity ^{51}V beam from the right-hand side bombards the rotating ^{248}Cm targets. The blue light is due to the luminescence emitted by He gas along the beam trajectory. The bright spot is due to the ^{248}Cm target area transited by the beam, with a size of approximately 8 mm$^{\mathrm{H}}$ $\times$ 1 mm$^{\mathrm{V}}$.

During about two years till the middle of 2022, 193 days were allocated for ^{248}Cm + ^{51}V measurement.

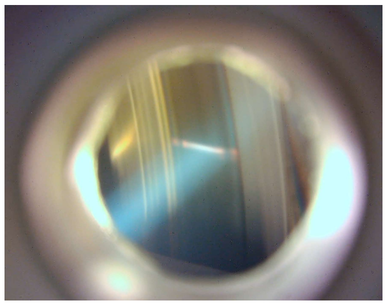

Fig. 9. Photograph of the ^{51}V beam crossing the target area at a rotating speed of 2,000 rpm.

7. Summary

This article described the "SHE project" initiated in 2016 at the RNC. The construction of SRILAC to boost the beam energy from 5.5 MeV/u to 6.5 MeV/u for rendering hot fusion reaction ^{51}V + ^{248}Cm possible to synthesize a new element (119) was one of the main objectives of the project.

During commissioning, which was completed in 2019, the performances of SRILAC and the newly built GARIS-III were confirmed. Thus, the initial objective of the SHE project was successfully achieved in its entirety.

The 119 search experiment is being carried out under an international collaboration called the nSHE[1] research group.

It is our great pleasure to acknowledge the invaluable contributions by B. Gall, R.K. Grzywacz, K. Morita, K. Rykaczewski, J.B. Roberto, S. Sakaguchi, E. Kako, K. Saito, N. Fukunishi, Y. Higurashi, Y. Watanabe, Z. Asfari, P. Brionnet, M. Fujimaki, H. Hasebe, S. Ishizawa, D. Kaji, S. Kimura, Y. Komori, A. Nambu, T. Niwase, Y. Shigekawa, M. Tanaka, T. Tanaka, F. Tokanai, Y. Wang, X. Yin, T. Yokokita, O. Kamigaito, H. En'yo, H. Sakurai and the nSHE Collaboration in the various phases. The ^{248}Cm isotope

[1] "n" could denote "n"ew, "n"ishina, or "n"ihon.

used in this research was supplied by the U.S. Department of Energy (DOE) isotope program, managed by the Office of Isotope R&D and Production. ORNL is managed by the UT-Battelle, LLC under DOE contract No. DE-AC05-00OR22725.

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