TASCA 26
22nd Workshop on Recoil Separator for Superheavy Element Chemistry & Physics

Gas phase chemical studies of the superheavy elements at the SHE Factory with a focus on Cn and Fl

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Joint Institute for Nuclear Research

SCIENCE BRINGING NATIONS TOGETHER

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SUPER HEAVY ELEMENT FACTORY
BASIC JINR FACILITY FOR SHE RESEARCH

2019

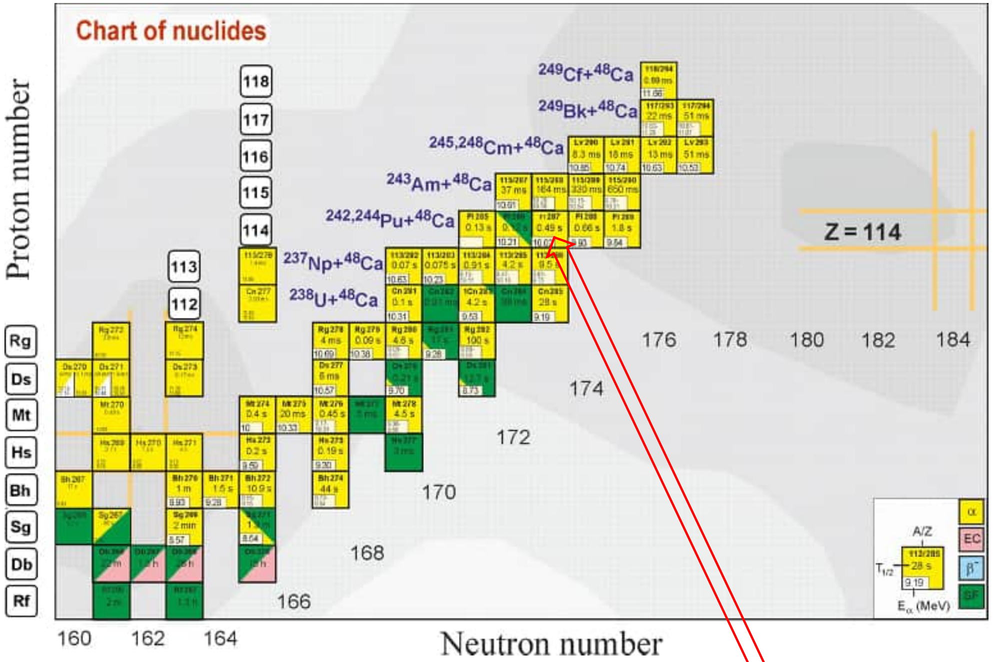
- Discovery of new SHE
- High statistics experiments:
  - nuclear chemistry
  - spectroscopy
  - mass measurements

2020

HI Cyclotron DC-280

Mission:
High intensity of heavy ion beams

SHE chemistry focus: isotopes produced in ^{48}Ca reactions




PHYSICAL REVIEW C **106**, 024612 (2022)

Investigation of ^{48}Ca-induced reactions with ^{242}Pu and ^{238}U targets at the JINR Superheavy Element Factory

Yu. Ts. Oganessian,[1] V. K. Utyonkov,[1] D. Ibadullayev,[1,2] F. Sh. Abdullin,[1] S. N. Dmitriev,[1] M. G. Itkis,[1] A. V. Karpov,[1] N. D. Kovrizhnykh,[1] D. A. Kuznetsov,[1] O. V. Petrushkin,[1] A. V. Podshibiakin,[1] A. N. Polyakov,[1] A. G. Popeko,[1] R. N. Sagaidak,[1] L. Schlattauer,[1,3] V. D. Shubin,[1] M. V. Shumeiko,[1] D. I. Solovyev,[1] Yu. S. Tsyganov,[1] A. A. Voinov,[1] V. G. Subbotin,[1] A. Yu. Bodrov,[1] A. V. Sabel'nikov,[1] A. Lindner,[1,3] K. P. Rykaczewski,[4] T. T. King,[4] J. B. Roberto,[4] N. T. Brewer,[4,5] R. K. Grzywacz,[4,5] Z. G. Gan,[6] Z. Y. Zhang,[6] M. H. Huang,[6] and H. B. Yang[1,6]
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Experiments using a ^{48}Ca beam on ^{238}U and ^{242}Pu targets to produce superheavy nuclei were perfor[...] the gas-filled separator DGFRS-2 online to the new cyclotron DC280 at the SHE Factory at JINR. The [...] properties of ^{286}Fl and ^{287}Fl, as well as their α-decay products, were refined after the detection of 25 and [...] decay chains, respectively. In addition, 16 decay chains of ^{283}Cn were observed in the ^{238}U + ^{48}Ca reacti[...] possibility of existing of isomeric states in the ^{287}Fl consecutive α decays is discussed. A new α line [...] energy of 100–200 keV lower than the main one at 10.19 MeV was observed for the first time for eve[...] ^{286}Fl decay. A maximum cross section of 10.4 pb was measured for the ^{242}Pu(^{48}Ca, 3n) ^{287}Fl reaction

First results:
DGFRS-2


PHYSICAL REVIEW C **106**, L031301 (2022)

First experiment at the Super Heavy Element Factory: High cross section of ^{288}Mc in the ^{243}Am + ^{48}Ca reaction and identification of the new isotope ^{264}Lr

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[3]Palacky University Olomouc, Department of Experimental Physics, Faculty of Science, 771 46 Olomouc, Czech Republic
[4]Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA
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(Received 11 March 2022; revised 2 May 2022; accepted 17 August 2022; published 29 September 2022)

We present results of the first experiment aimed at the synthesis of Mc isotopes in the ^{243}Am + ^{48}Ca reaction performed at the new gas-filled separator DGFRS-2 online to the new cyclotron DC280 at the Super Heavy Element Factory at JINR. Fifty-five new decay chains of ^{288}Mc and six chains assigned to ^{289}Mc were detected. The α decay of ^{268}Db with an energy of 7.6–8.0 MeV, half-life of 16^{+6}_{-4} h, and a branch of 55^{+20}_{-15}% was registered for the first time, and a new spontaneously fissioning isotope ^{264}Lr with a half-life of $4.9^{+2.1}_{-1.3}$ h was identified. The cross section for the ^{243}Am(^{48}Ca, 3n) ^{288}Mc reaction was measured to be $17.1^{+6.3}_{-4.7}$ pb, which is the largest value for the known superheavy nuclei at the island of stability.

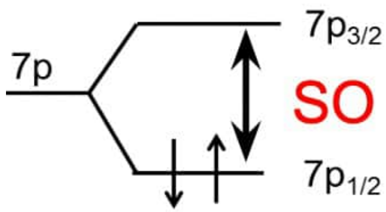
Higher statistics
New data

The chemistry of Fl is the most exciting!

Discovered in 1999

Periodic trends down the group:

- +2 oxidation state
- Increasing the stability of elem. states
- Enhancement of metallic properties
- The properties in group 14 differ more from each other than in other groups



Spin-Orbit stabilization of Fl atom makes it much less reactive than Pb

Experimental confirmation of the strong influence of relativistic effects is needed!

B. Eichler, 1974
High volatility of elements 112-118

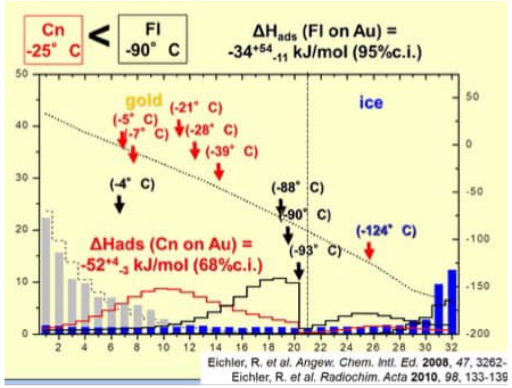
Pitzer, 1975: 112 and 114 elements will be very inert, like noble gases

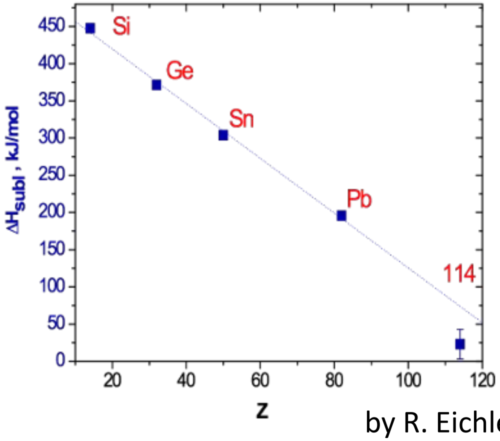
Led to larger number of modern calculations and model experiments FLNR, PSI, GSI etc. ⇒

Gas adsorption thermochromatography on gold method developed to study one atom behavior

Fl chemistry, several runs in two labs, 2006-2015

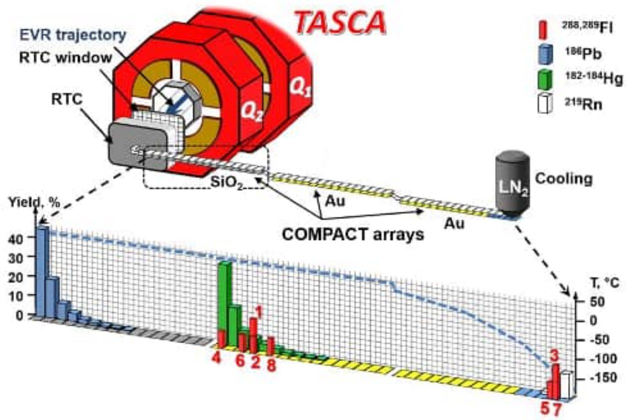
COLD setup, PSI behind the target@U-400, FLNR





by R. Eichler

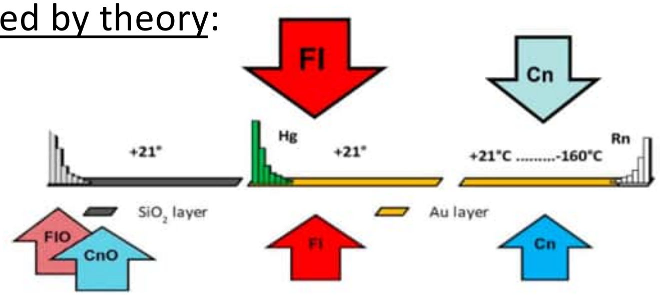
COMPACT setup behind the TASCA separator, GSI



Yakushev A. et al. On the adsorption and reactivity of element 114, flerovium // Front. Chem. 2022, 10:976635.

- **Two deposition zones observed! Questions remain:**
- High volatility and inert behavior - in elemental state?
- Physi- or chemisorption on gold?
- Very volatile inert metal or gas?

Supported by theory:



by V. Pershina

Further research with higher statistics is needed!

SHE Factory

New separator at SHE Factory

Gasfilled Recoil Analyzer and Nuclei Detector – GRAND

A.V. Yeremin, et al.
Universal GRAND Gas-Filled Separator: First Experimental Results.
PEPAN Letters, Vol. 21, No. 3, pp. 518–525 (2024)

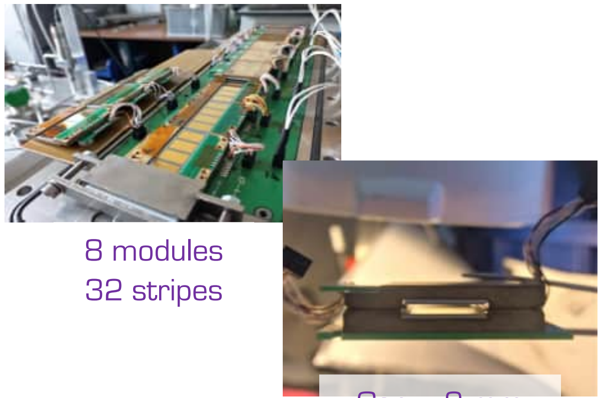
Launched in 2022

Alexander SVIRIKHIN
Spectroscopy of heaviest nuclei

- DGFRS-2 analogue
- Helium
- Transmission 50 %
- Two setups
- Joint team
  chemists and physicists

Cryodetector setup for Nh chemistry, FLNR

Detection system



ISSN 0020-4412, Instruments and Experimental Techniques, 2010, Vol. 53, No. 6, pp. 789–791. © Pleiades Publishing, Ltd., 2010.
Original Russian Text © A.V. Isaev, A.V. Yeremin, N.I. Zamyatin, E.V. Zubarev, A.N. Kuznetsov, O.N. Malyshev, O. V. Petrushkin, A.V. Sabel'nikov, A.I. Svirikhin, M.L. Chelnokov, V.I. Chepigin, S.N. Dmitriev, 2010, published in Pribory i Tekhnika Eksperimenta, 2010, No. 6, pp. 16–20.

NUCLEAR EXPERIMENTAL TECHNIQUE

Detecting System of the Setup for Studying Chemical Properties of Superheavy Elements Using the Gas Chemistry Techniques

A. V. Isaev[a*], A. V. Yeremin[a], N. I. Zamyatin[b], E. V. Zubarev[b], A. N. Kuznetsov[a], O. N. Malyshev[a], O. V. Petrushkin[a], A. V. Sabel'nikov[a], A. I. Svirikhin[a], M. L. Chelnokov[a], V. I. Chepigin[a], and S. N. Dmitriev[a]
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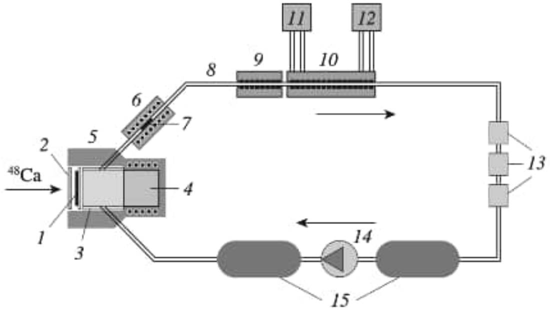
Received March 31, 2010

U-400: behind the target



ELSEVIER
Available online at www.sciencedirect.com
ScienceDirect
Mendeleev Commun., 2014, 24, 253–256
Mendeleev Communications

Pioneering experiments on the chemical properties of element 113

Sergey N. Dmitriev,[a*] Nikolay V. Aksenov,[a] Yuriy V. Albin,[a] Gospodin A. Bozhikov,[a] Maxim L. Chelnokov,[a] Viktor I. Chepygin,[a] Robert Eichler,[b] Andrei V. Isaev,[a] Denis E. Katrasev,[a] Vyacheslav Ya. Lebedev,[a] Oleg N. Malyshev,[a] Oleg V. Petrushkin,[a] Lidia S. Porobanuk,[a] Mikhail A. Ryabinin,[c] Alexey V. Sabel'nikov,[a] Evgeny A. Sokol,[a] Alexander V. Svirikhin,[a] Gennadii Ya. Starodub,[a] Ilya Usoltsev,[b] Grigory K. Vostokin[a] and Alexander V. Yeremin[a]
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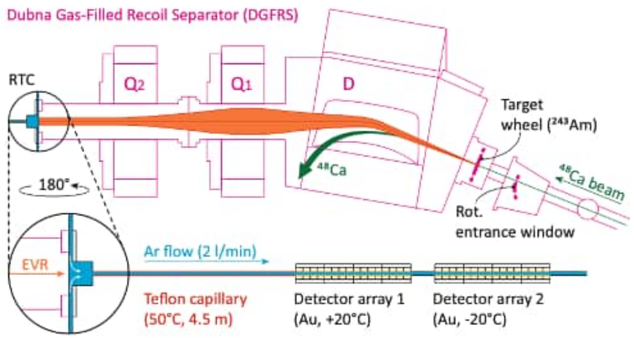
DOI: 10.1016/j.mencom.2014.09.001

First experimental results of a chemical investigation of element 113 independently confirm the synthesis of the new elements 115 and 113 in the nuclear fusion reaction of ^{48}Ca + ^{243}Am and indicate a chemical behaviour resembling a species with a high volatility, a weak interaction with inert surfaces and an enhanced reactivity towards gold surfaces.

U-400: behind the DGFRS



Eur. Phys. J. A (2017) 53: 158
DOI 10.1140/epja/i2017-12348-8
THE EUROPEAN PHYSICAL JOURNAL A
Regular Article – Experimental Physics

On the volatility of nihonium (Nh, Z = 113)

Nikolay V. Aksenov[1], Patrick Steinegger[1,a], Farid Sh. Abdullin[1], Yury V. Albin[1], Gospodin A. Bozhikov[1,2], Viktor I. Chepigin[1], Robert Eichler[3,4], Vyacheslav Ya. Lebedev[1], Alexander Sh. Madumarov[1], Oleg N. Malyshev[1], Oleg V. Petrushkin[1], Alexander N. Polyakov[1], Yury A. Popov[1], Alexey V. Sabel'nikov[1], Roman N. Sagaidak[1], Igor V. Shirokovsky[1], Maksim V. Shumeiko[1], Gennadii Ya. Starodub[1], Yury S. Tsyganov[1], Vladimir K. Utyonkov[1], Alexey A. Voinov[1], Grigory K. Vostokin[1], Alexander V. Yeremin[1], and Sergey N. Dmitriev[1]
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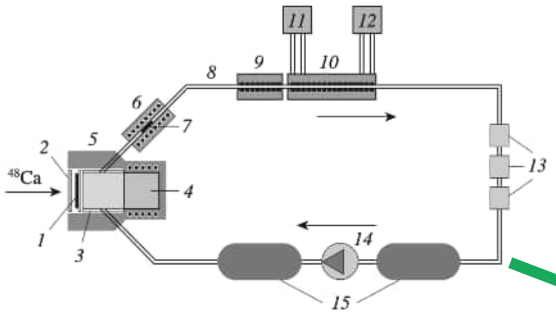
[4] Universität Bern, Departement für Chemie und Biochemie, Freiestrasse 3, 3012 Bern, Switzerland

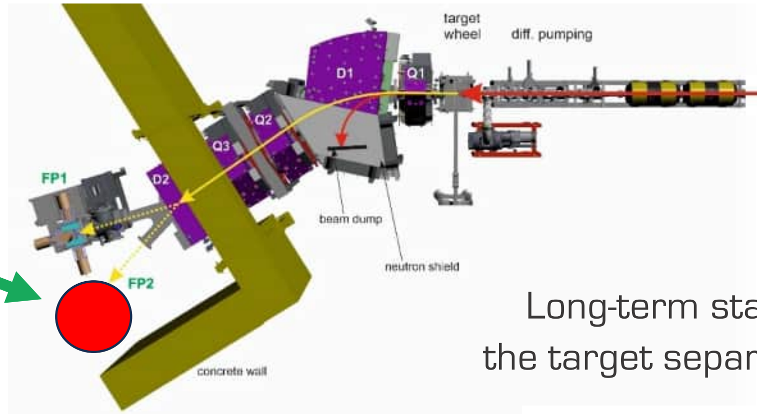
Cryodetector setup to couple to GRAND separator, DC-280

Cryodetector setup

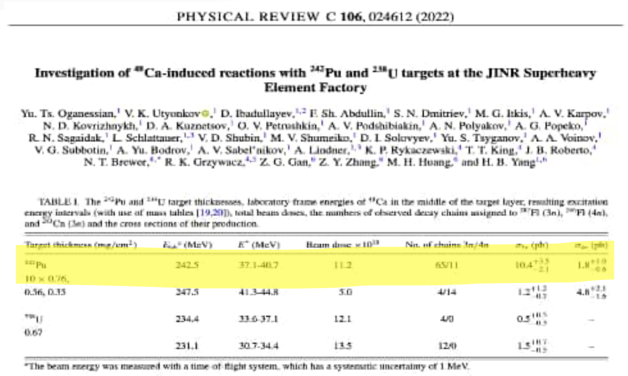


New GRAND separator

DC-280 Cyclotron



Long-term stable **5 pμA** of ^{48}Ca on the target separator with **stable** energy



PHYSICAL REVIEW C **106**, 024612 (2022)

Investigation of ^{48}Ca-induced reactions with ^{242}Pu and ^{238}U targets at the JINR Superheavy Element Factory

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TABLE I. The ^{242}Pu and ^{238}U target thicknesses, laboratory frame energies of ^{48}Ca in the middle of the target layer, resulting excitation energy intervals (with use of mass tables [19,20]), total beam doses, the numbers of observed decay chains assigned to ^{287}Fl (3n), ^{286}Fl (4n), and ^{283}Cn (3n) and the cross sections of their production.

Target thickness (mg/cm^2)	E_{lab}[a] (MeV)	E^* (MeV)	Beam dose $\times 10^{18}$	No. of chains 3n/4n	σ_{3n} (pb)	σ_{4n} (pb)
^{242}Pu	242.5	37.1-40.7	11.2	65/11	$10.4^{+3.5}_{-2.1}$	$1.8^{+1.0}_{-0.6}$
10 × 0.76,						
0.56, 0.33	247.5	41.3-44.8	5.0	4/14	$1.2^{+1.2}_{-0.7}$	$4.8^{+2.1}_{-1.6}$
^{238}U	234.4	33.6-37.1	12.1	4/0	$0.5^{+0.5}_{-0.3}$	–
0.67						
	231.1	30.7-34.4	13.5	12/0	$1.5^{+0.7}_{-0.5}$	–

[a]The beam energy was measured with a time of flight system, which has a systematic uncertainty of 1 MeV.

1. **To rebuild FLNR setup for Cn and Fl chemistry**
2. **To reproduce PSI and/or GSI experiments**
3. **Speciation w/surface interaction study**

BEAM TIME $^{242}Pu(^{48}Ca, 3n)^{287}Fl$

Joint team of chemists and physicists (Spectroscopy sector by A. Svirikhin)

Run 1. December 2022:

Target – 240 mm, 0.7 mg/cm^2

Mean beam intensity – 2 μA $^{48}Ca^{10+}$

Beam integral –1.6×10^{19}

29 days

Setup in "Nh-mode"

Old RTC

Gas flow 2 L/min

Capillary 40 cm

He/Ar = 60/40, 1,3 bar

Temperature gradient 20 °C - –170 °C

Transfer from U-400 to SHE Factory: Pre-separation approach

What do we need from a separator?

- Rotating target - Increase beam and production rate
- Separation from by-product actinides (alpha, SF) – low background at Cryodetector
- Separation from ^{48}Ca beam – «Clean» conditions in gas phase
- Setup placement at the focal plane – faster gas transport

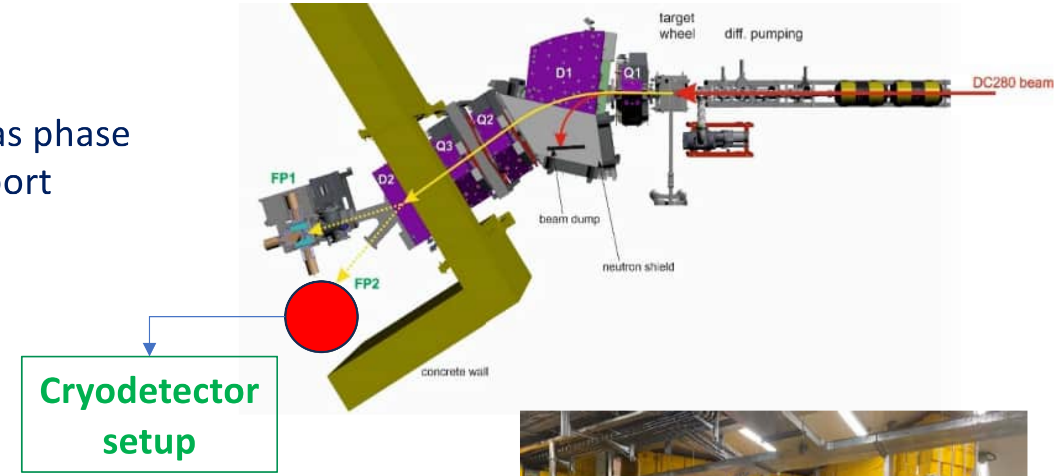
Disadvantages of GRAND for chemistry

- Decrease isolation efficiency (at least x2)
- Pressure difference between setups – gas leak into separator
- **Large focal image - increase transport time**

To overcome: Development of high-speed recoil collection gas cell

- ❑ Safe connection between Cryodetector and GRAND
- ❑ Gas transport time and efficiency to detector array must be fast enough to effectively study the ^{287}Fl with a half-life of 360 ms

GRAND separator as a pre-separator tool



Experimental hall

Research at GRAND in preparation for Fl chemistry (2023-2024) to be published

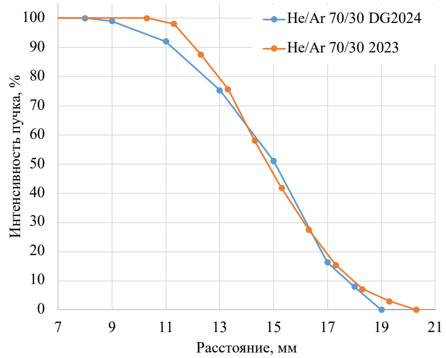
Joint team of chemists and physicists (Spectroscopy sector by A. Svirikhin) from FLNR and IMP CAS, China

1. Stopping range measurements
   - ✓ Mylar thickness
   - ✓ Double grid efficiency
   - ✓ He/Ar mixtures

Test reactions:

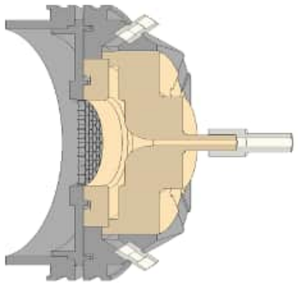
$^{208}Pb(^{48}Ca, 2n)^{254}No$ ($E_{ER} \approx 40$ MeV)

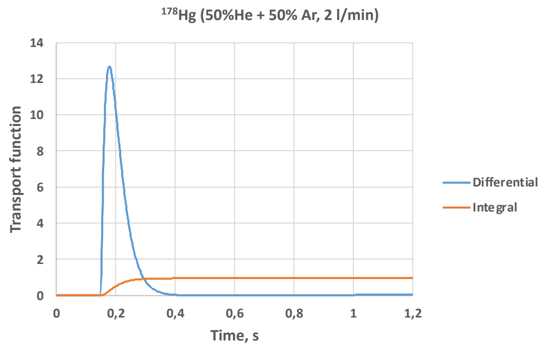
$^{144}Sm(^{40}Ar, xn)^{184-n}Hg$ ($E_{ER} \approx 47$ MeV)



2. Modeling: FP recoil collection effective area vs $T_{1/2}$ Fl-287
3. Gas flow modeling
4. Development and manufacture

5. On-line measurements using Cryodetector setup



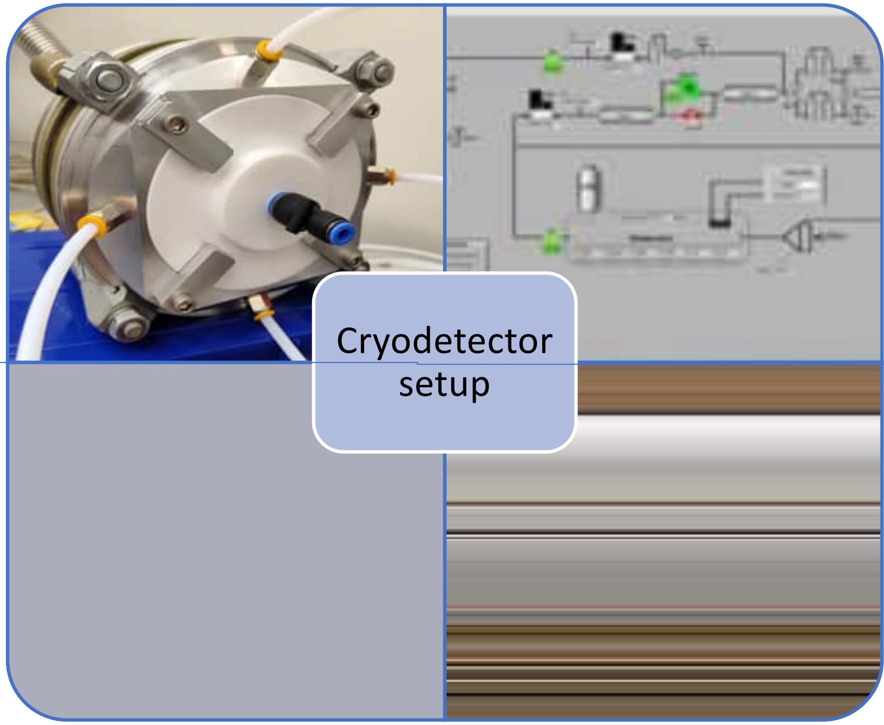


- ✓ **The transport efficiency of the new cell is 3-5 times higher**
- ✓ **Mean transport time measured – 200 ms, 2 L/min, 50/50 = He/Ar**

Cryodetector setup successfully rebuilt for Fl chemistry (2024) to be published

- **New RTC**
- **Speed and efficiency performance improved**

- **Gas loop system improvement**
- **New gas composition and leaking control**



- **New Au coated detectors**
- **Detection systems improvement**

- **New LN cooling system**
- **Temperature gradient stability tests**

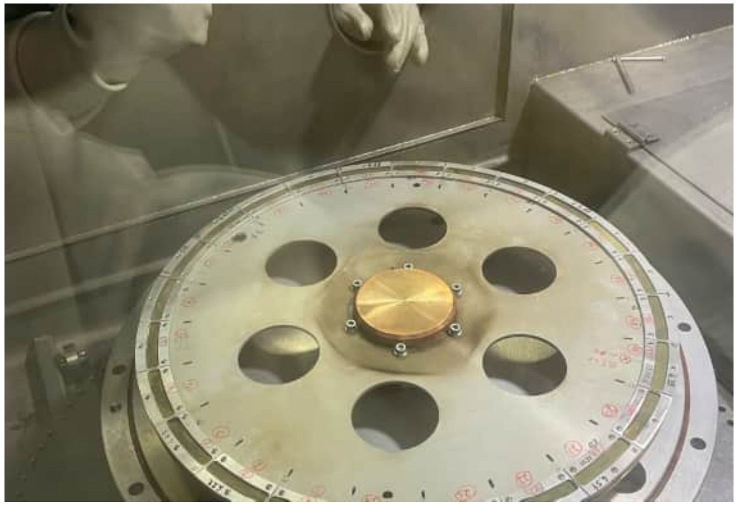
- on-line research with short-lived 200 ms ^{178}Hg
- on-line, non-stop, reliable for 30 days run
- Etc.

The 480 mm diameter target on GRAND

- **Year: 2025**
- **$^{242}Pu(^{48}Ca, 3\text{-}4n)^{287\text{-}286}Fl$**
- PuO_2 target: 697±50 μg/cm^2 on 1.5 μm Ti backing
- Wheel diameter: 480 mm
- Total area: 141 cm^2
- Segments: 24
- **Beam: 5 pμA**
- Gas on separator: He

the target is stable during several weeks of irradiation without any significant loss of the material!

Data analysis is in progress…



Alexey Sabelnikov and Alexander Bodrov

Target module

O.N. Malyshev, E. Teymurov and Yu. A. Popov

BEAM TIME $^{242}Pu(^{48}Ca, 3n)^{287}Fl$

Joint team of chemists and physicists (Spectroscopy sector by A. Svirikhin) from FLNR and IMP CAS, China

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Mean beam intensity – 2 μA $^{48}Ca^{10+}$

Beam integral –1.6×10^{19}

29 days

Setup in ”Nh-mode”

Old RTC

Gas flow 2 L/min

Capillary 40 cm

He/Ar = 60/40, 1,3 bar

Temperature gradient 20 °C - −170 °C

CRYODETECTOR IMPROVEMENT

Run 2. June 2025:

Target – 480 mm, 0.7 mg/cm^2

Mean beam intensity up to 5 μA $^{48}Ca^{10+}$

Beam integral – 3.4×10^{19}

Fl synthesis/separation efficiency decreased

New RTC

Gas flow 1,5 L/min

Capillary 40 cm

He/Ar = 60/40, 1,3 bar

Temperature gradient 20 °C - −170 °C

Dew point –109 °C

GRAND as a preseparator for chemistry

pros:

- Rotating target
- Separation from ^{48}Ca beam
- Separation from actinides (alpha+SF)
- «Clean» conditions for chemistry

cons:

- less efficiency (at least x2)
- large focal image
- Pressure drop

2025

Separator efficiency

droped x4-6

😢

more beam diagnostic

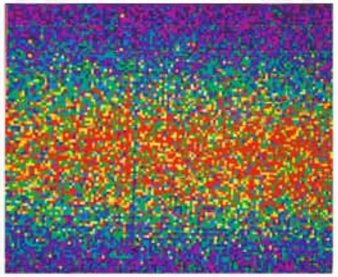
tools development are needed…

Tests of ion optics

$^{48}Ca + ^{170}Er \rightarrow 4n + ^{214}Ra$

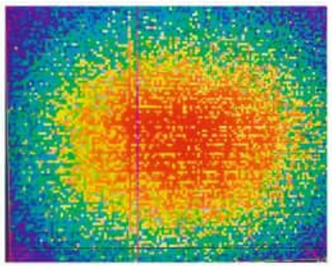
FP1 – 100x100 cm^2 DSSD

Vertical distribution



Horizontal distribution

$Q_v D_{30} Q_h Q_v D_{15}$



Horizontal distribution

$Q_v D_{30} Q_v Q_h D_{15}$

Separator **GRAND**

Gas-filled **R**ecoil **A**nalyzer and **N**uclei **D**etector

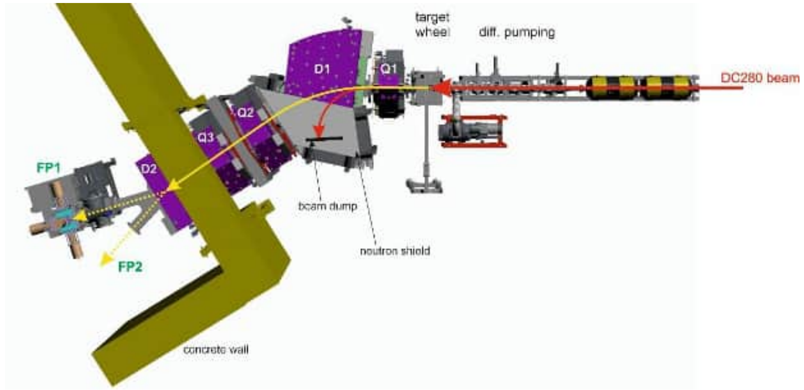


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Table 1. Comparative parameters of gas-filled separators used in experiments on the synthesis and study of the properties of isotopes of heavy elements

Facility	Configuration	Acceptance angle, msr	Rotation angle	Max. Bρ, T m	Dispersion mm/% Bρ	Length, m	Ref.
DGFRS1	$D_vQ_hQ_v$	10	23°	3.1	7.5	4.07	[5]
DGFRS2	$Q_vD_hQ_hQ_vD$	16	(32 + 10)°	3.35	32.8	7.41	[6]
BGS	Q_vD_hD	45	(25 + 45)°	2.5	20.0	4.6	[7]
GARIS1	$D_{vh}Q_hQ_vD$	12.2	(45 + 10)°	2.16	9.7	5.76	[8]
GARIS2	$Q_vD_hQ_hQ_vD$	18.5	(30 + 7)°	2.43	19.3	5.06	[9]
TASCA	DQ_hQ_v	13.3	30°	2.4	9.0	3.5	[10]
RITU	$Q_vDQ_hQ_v$	8.5	25°	2.2	10.0	4.8	[11]
SHANS	$Q_vDQ_vQ_h$	25	52°	2.88	7.3	6.5	[12]
SHANS2	$Q_vDQ_hQ_vD$	25	(30 + 10)°	2.56	20.9	5.97	[13]
AGFA	Q_vD_h	44–22[a]	38°	2.5		3.7–4.3[a]	[14]
GRAND	$Q_vD_hQ_vQ_hD$	16	(32 ± 15)°	3.35	32.8	7.95[b]; 6.84[c]	This work

[a] Distance to target up to Q 40 cm or 84 cm, [b] distance from the target to the "physical" focal detector, and [c] distance from the target to the "chemical" stop chamber of RNs.

Preliminary Results: 8 decay chains detected in the Cryodetector (to be published)

$^{48}Ca + ^{242}Pu \rightarrow$ ^{287}Fl ($T_{1/2}$ = 0.36 s) $\rightarrow$ ^{283}Cn ($T_{1/2}$ = 3.81 s)

*

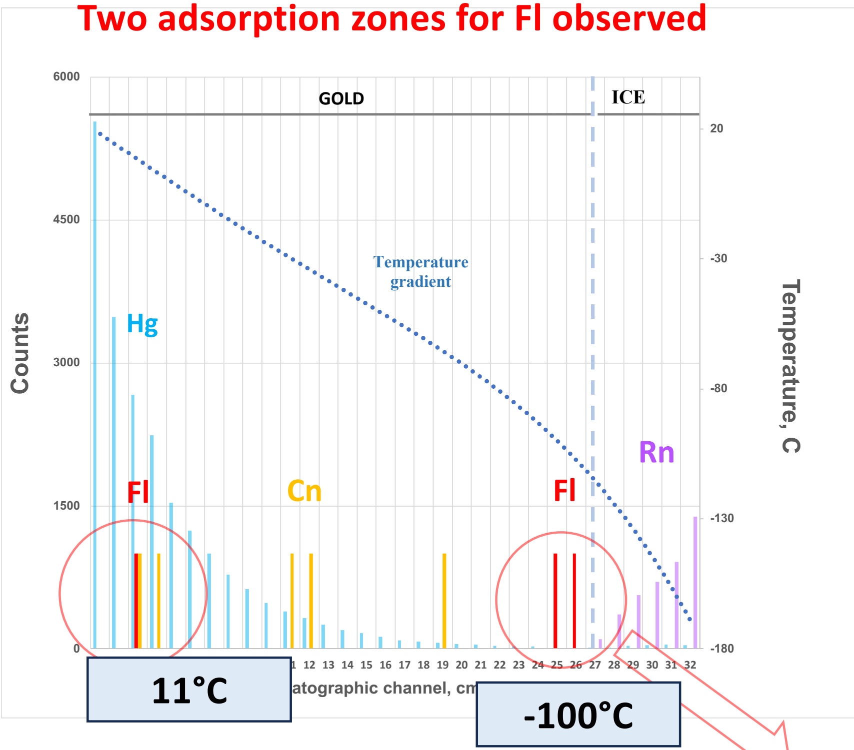
^{287}Fl 360^{+45}_{-36} ms α 10,016	^{287}Fl α 9,76	^{287}Fl α 9,8	^{287}Fl FF 60+86	^{287}Fl missing	^{287}Fl missing	^{287}Fl missing	^{287}Fl missing	^{287}Fl missing
^{283}Cn $3,81^{+0,45}_{-0,36}$ s α 9,531	^{283}Cn 1,3 s α 6.19—6,58	^{283}Cn missing		^{283}Cn α 9,48	^{283}Cn α 9,07*	^{283}Cn α 9,52	^{283}Cn α 9,31	^{283}Cn α 9,52
^{279}Ds 186^{+21}_{-17} ms α 9,686 SF	^{279}Ds 0,38-0,82 s FF 110+missing	^{279}Ds 5,43 s FF 86+84		^{279}Ds 0.41 s FF 91+80	^{279}Ds 0,56 s FF79+4**	^{279}Ds 0,13 s FF 80+68	^{279}Ds 0.41 s FF 80+40	^{279}Ds 0,53 s FF 60+86
	-100°C	11°C	-100°C	-19°C	7°C	-22°C	-70°C	11°C

3/3 of ^{287}Fl 1-3 of ^{283}Cn are probably missing   No SF-background

* *Oganessian Yu.Ts. et al.* Investigation of ^{48}Ca-induced reactions with ^{242}Pu and ^{238}U targets at the JINR Superheavy Element Factory // Phys. Rev. C. 2022, 106, 024612.

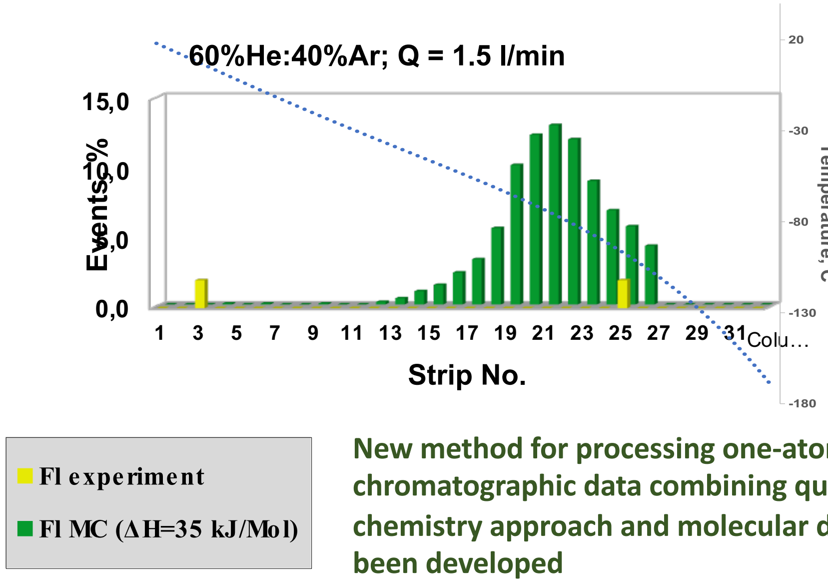
THERMOCHROMATOGRAPHY OF Fl ON GOLD IN INERT GAS FLOW

Two adsorption zones for Fl observed



Further speciation study is needed!

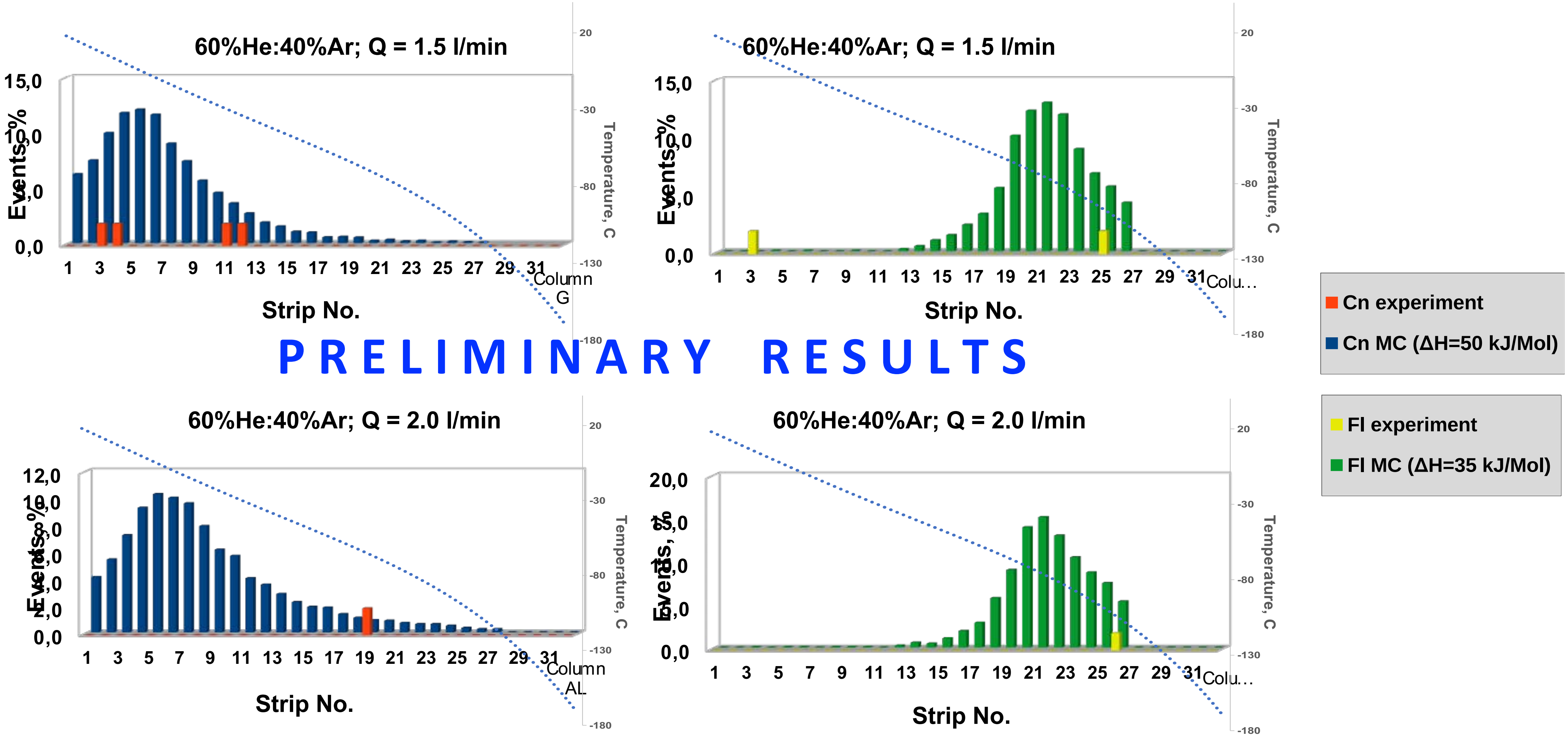
New data in sum with data from PSI and GSI detectors is sufficient to draw a conclusion



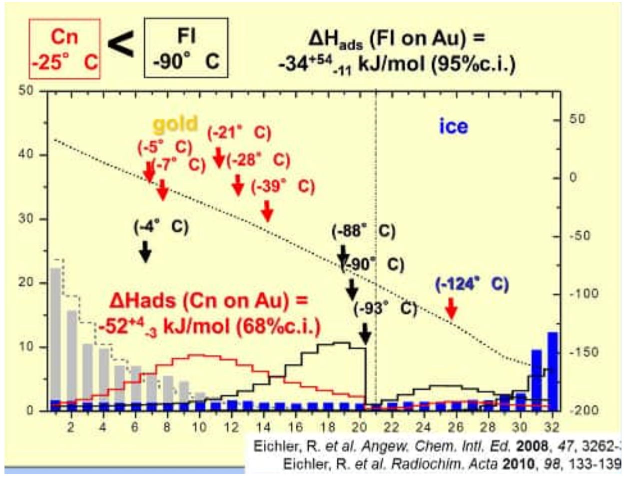
New method for processing one-atom chromatographic data combining quantum chemistry approach and molecular dynamics has been developed

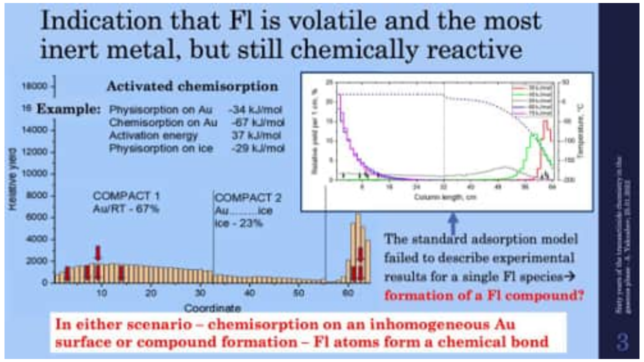
No more statistics is needed in this system

PRELIMINARY RESULTS

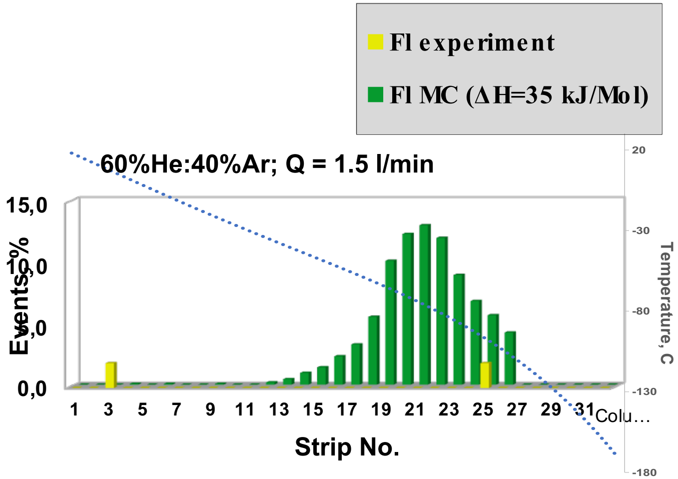


COLD | **COMPACT** | **Cryodetector**





From A. Yakushev



Under discussion and calculation

1. Formation of two chemical forms Fl and FlO

- FlO formation and stability theoretically confirmed, FlH - not
- possible reaction in the gas with O_2 traces – can be studied
- possible reaction on the gold surface – can be studied

2. Adsorption on inhomogeneous gold surface + activation barrier

- Mechanism not clear but two different bound states

3. Cn adsorption on polycrystalline gold 111 + 200, Fl – not

4. Low statistics: surface contamination and decay in flight

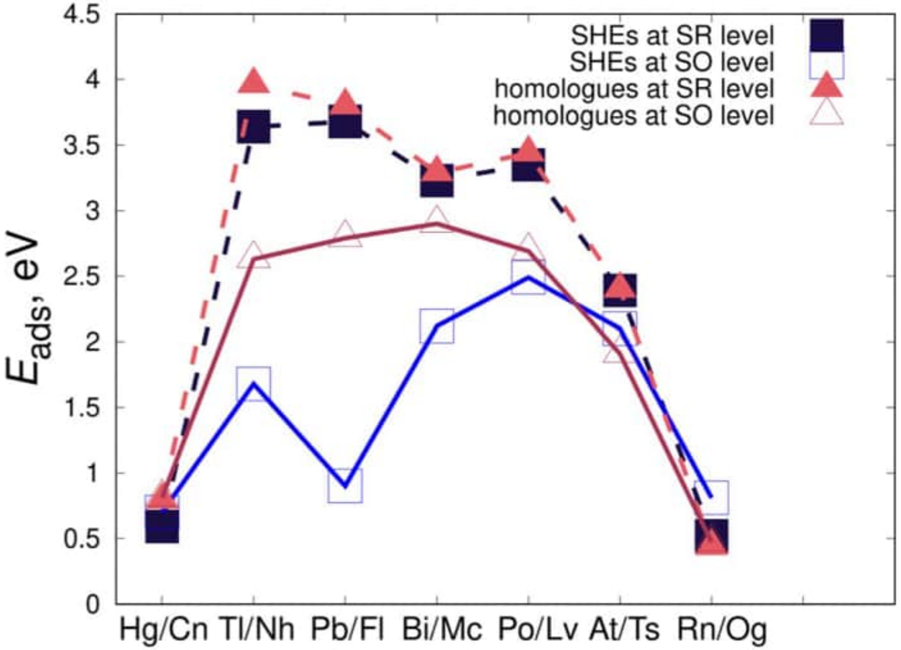
✓ **Rebuild FLNR setup for Fl chemistry**

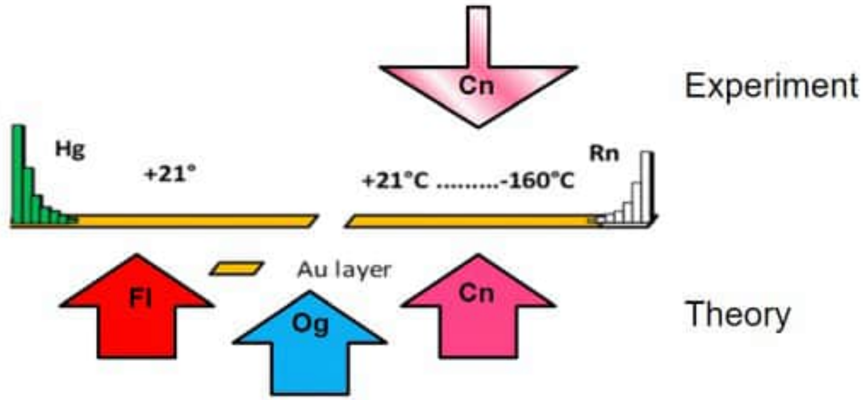
✓ **Reproduce PSI and/or GSI experiments**

❑ **Speciation study**

- ❑ + H_2
- ❑ + high temp. getter
- ❑ + O_2

Theory

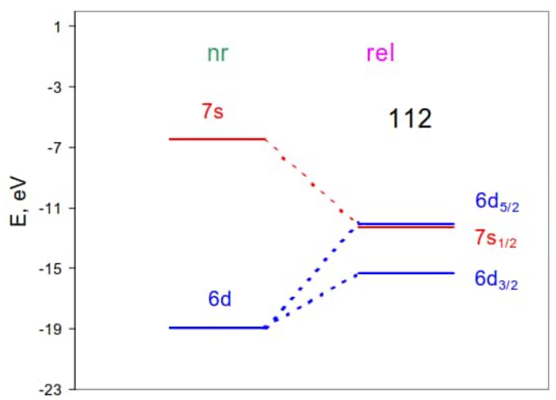




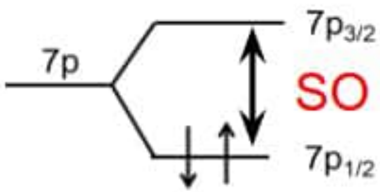
A. Ryzhkov et al. Phys. Chem. Chem. Phys. 2023, 25, 15362

E_{ads}(Fl) = 0.40 – 0.90 eV

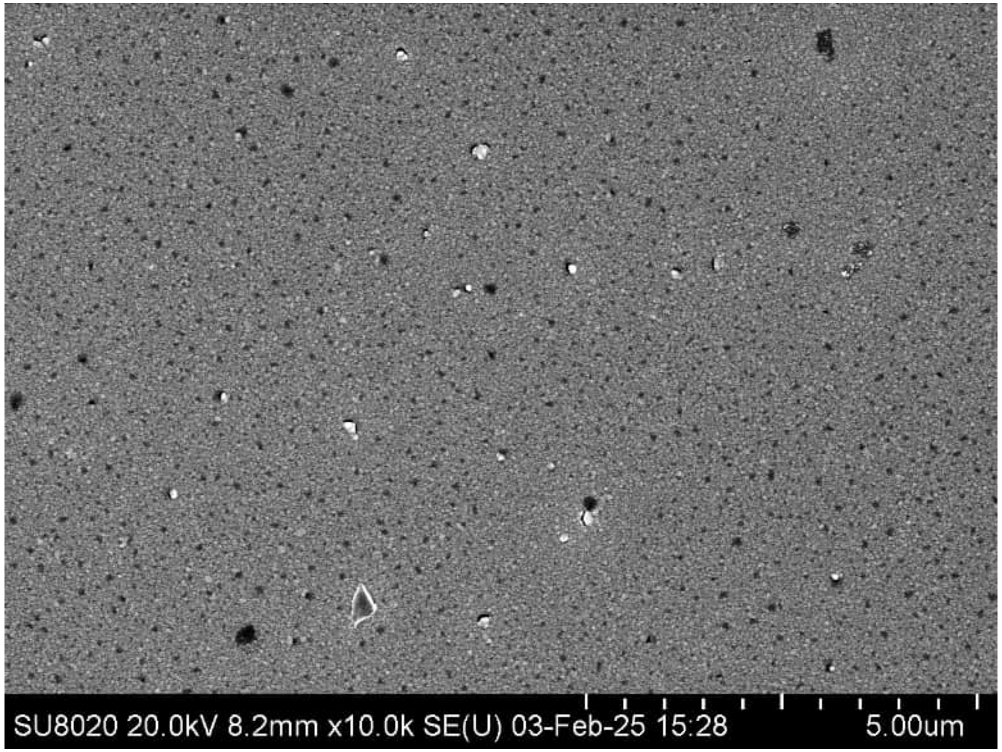
E_{ads}(Cn) = 0.35 – 0.80 eV



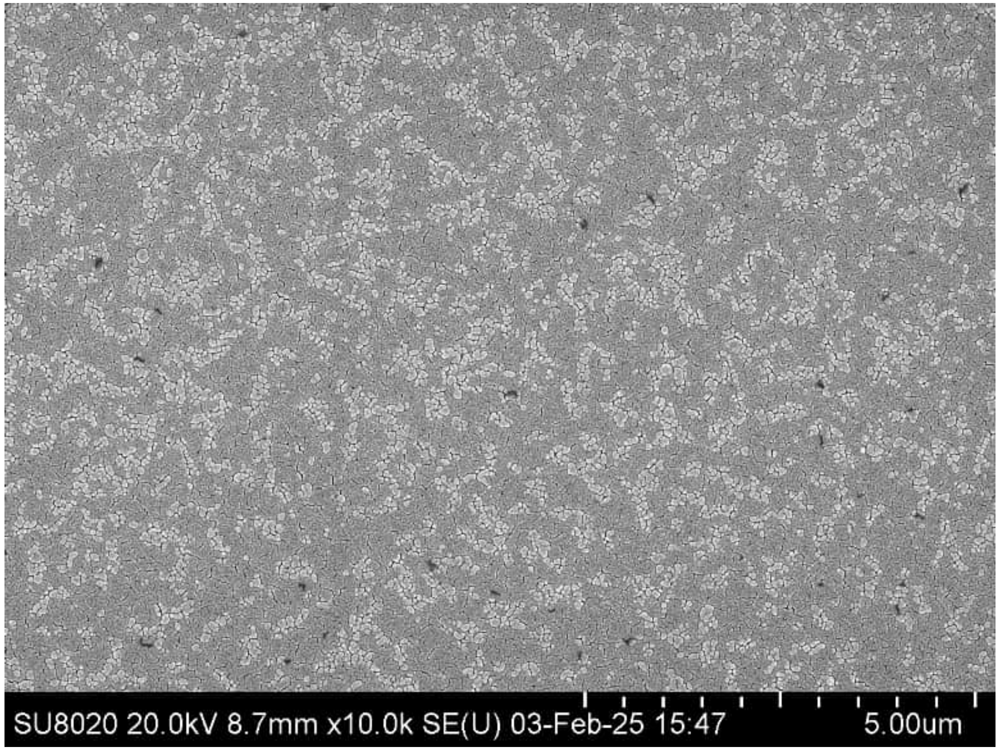
Pb: -1.88 eV
Fl: -5.12 eV



SEM

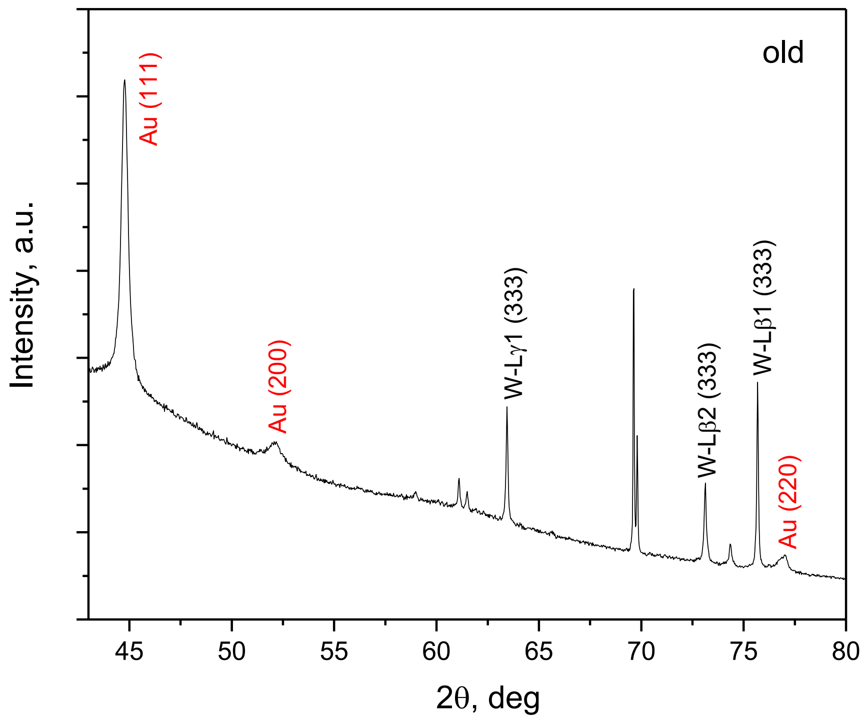


Old detection module

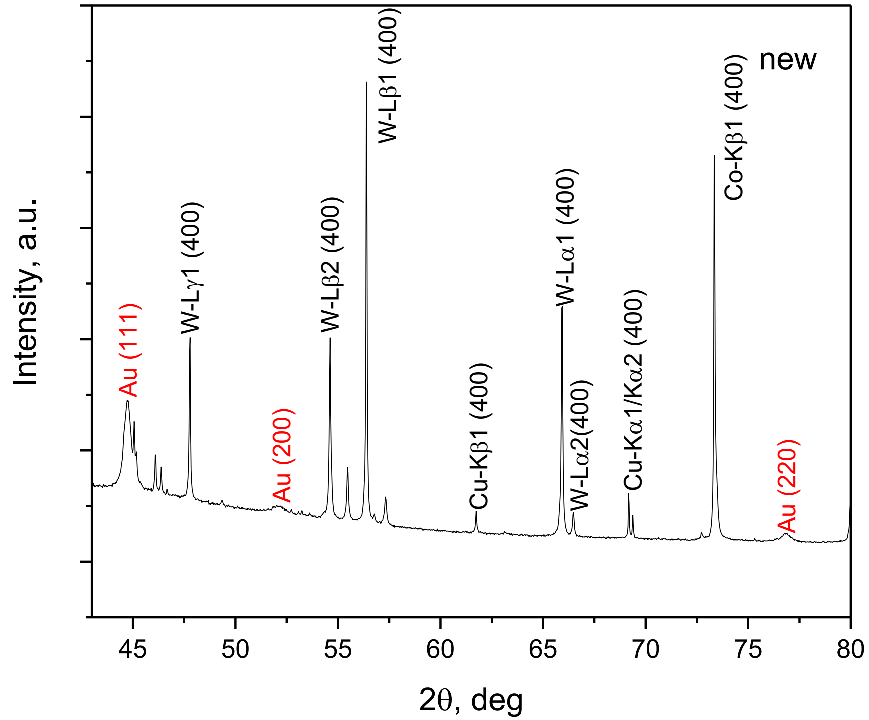


New detection module

XRD

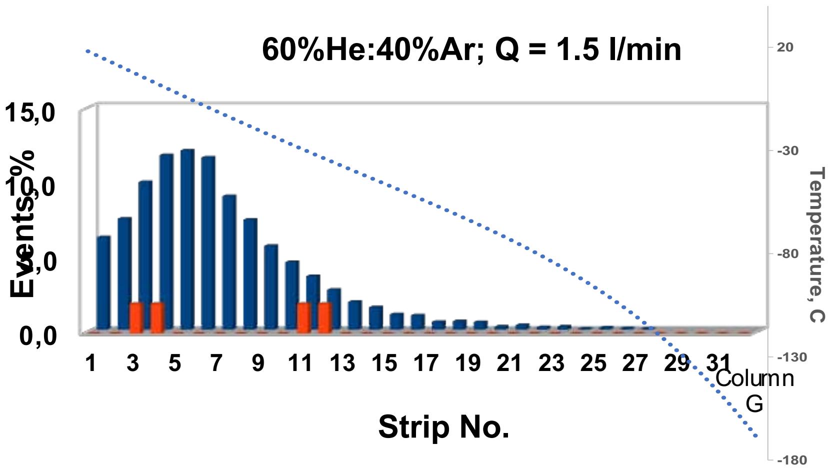


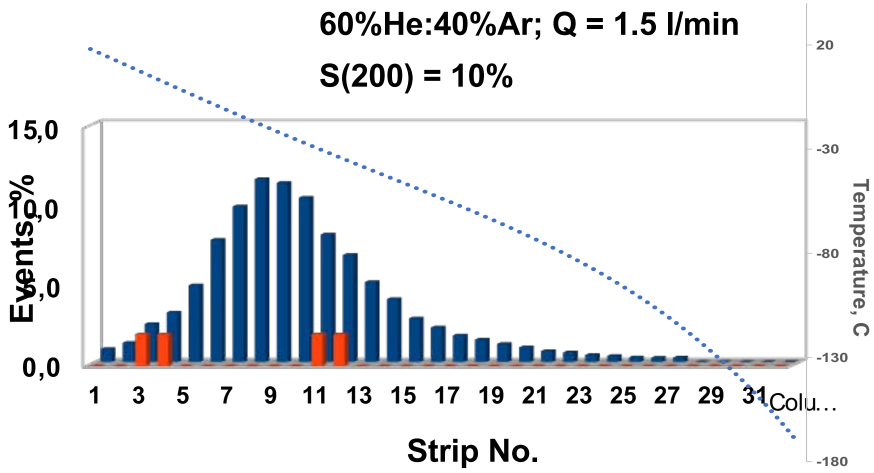
Old detection module



New detection module

Surfaces (111) and (200)



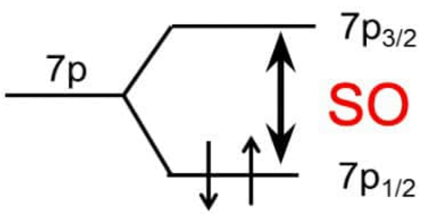


First step of the GASSOL radiochemistry research program

How strong was the observed influence of relativistic effects?
Why do we observe two adsorption temperatures and how can this be studied?

Speciation study is needed, a chemical sensitive and selective approach in 3 experiments:

- ❑ stabilization of elemental Fl in a reductive gas
- ❑ thermal decomposition of Fl oxide
- ❑ Fl oxide formation in surface reactions with oxygen



Experimentally observed influence of relativistic effects:
Spin-Orbit stabilization of Fl atom makes it much less reactive than Pb

Summary

- New data on Fl chemical properties in gas phase were obtained
- Next Fl chemistry are planned at the new dedicated for SHE chemistry separator GASSOL in 2028
- New setup but more theory first…

CHEMISTRY AT THE SUPERHEAVY ELEMENT FACTORY

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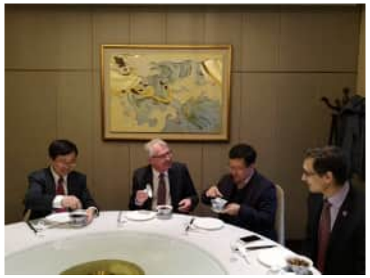

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