SYNTHESIS AND STUDY OF PROPERTIES
of SUPERHEAVY ELEMENTS AT FLNR.
PRESENT STATUS AND FUTURE

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United Nations Educational, Scientific and Cultural Organization
International Union of Pure and Applied Chemistry

“Chemistry – our life, our future”

The 100th anniversary of the Nobel Prize in Chemistry awarded to Marie Skłodowska-Curie
Synthesis of SHE via hot fusion of heavy target nuclei with light projectiles

- Rutherfordium (Z=104)
- Nobelium (Z=102)
- Lawrencium (Z=103)
- Seaborgium (Z=106)
- Dubnium (Z=105)

By irradiation of actinides with light ions in Berkeley (USA) and Dubna (Russia) the elements up to Z=106 could be synthesized.

The linear accelerator UNILAC and the velocity filter SHIP at GSI allowed for the synthesis of elements with Z=107-112.

Synthesis of SHE via cold fusion (Pb and Bi as target nuclei)

- Element 112
- Element 113

The linear accelerator UNILAC and the velocity filter SHIP at GSI allowed for the synthesis of elements with Z=107-112.
Cold & hot fusion cross sections

- Cold fusion: $^{208}\text{Pb, }^{209}\text{Bi + }^{48}\text{Ca, }^{50}\text{Ti, ... }^{70}\text{Zn}$
- Hot fusion: Actinides + $^{22}\text{Ne, }^{26}\text{Mg, ... }^{34}\text{S}$

Cross sections (cm$^2$)

Atomic number

SHE
Efforts focused on the synthesis of SHE

**NEW ECR-ion source**  
(GANIL, JINR)

**ACCELERATORS**

- beam intensity - $4 \times 8 \times 10^{12} / s$
- beam time - 4000 h/y

**ISOTOPE ENRICHMENT**

- Isotopes: $^{233, 238}$U, $^{242, 244}$Pu, $^{243}$Am, $^{245, 248}$Cm, $^{249}$Cf
- $^{48}$Ca (Lesnoy)
- Enrichment up to 68-70%
- Isotope production in high flux reactors (Oak Ridge, Dimitrovgrad)

**TARGET TECHNOLOGY**

- Technology of the target preparation - 0.3 mg/cm²
- Isotope enrichment 98-99%
- S-2 separator (Sarov)

**NEW TARGET MATTER**

- New target matter technology

**SEPARATION AND DETECTION**

- Separation and detection of superheavy nuclei
- New separator & detectors (Dubna, Livermore)

**NEW RECOIL SEPARATOR**

- New ECR-ion source (GANIL, JINR)

**NEW REACTOR REGIME**

- Reactor regime

- Efforts focused on the synthesis of SHE
FLNR U400 cyclotron
Number of observed decay chains
Element 118       3
Element 116     26
Element 115       4
Element 114     43
Element 113       2
Element 112       8
Cold & hot fusion cross sections

Cross sections (cm$^2$)

1n □ Cold fusion
$^{208}$Pb, $^{209}$Bi + $^{48}$Ca, $^{50}$Ti,... $^{70}$Zn

4n ○ Hot fusion
Actinides + $^{22}$Ne, $^{26}$Mg,... $^{34}$S

4n-3n

Actinides + $^{48}$Ca

survival

SHE

Atomic number
The Bk-249 was produced at ORNL (USA) by irradiation: of Cm and Am targets for approximately **250 days** by thermal-neutron flux of **$2.5 \times 10^{15}$ neutrons/cm$^2\cdot$s** in the HFIR (High Flux Isotope Reactor).
249Bk was produced at High Flux Reactor in Oak-Ridge.............March 09, 2009

22 mg of pure 249Bk was shipped to Moscow..........................June 20, 2009
Arrived at Dimitrovgrad (Russia) for target preparation...............July 01, 2009
Experiment have started at Dubna...........................................July 27, 2009
Experimental Setup

$v (A=48) = 0.11 \, c$
$q = 16.5^+$

$v (A=288) = 0.017 \, c$
$q = 6.2^+$

Dubna Gas Filled Recoil Separator
Target

$\omega = 1700 \text{ rpm}$

$6 \text{ cm}^2$

$310 \mu\text{g/cm}^2 \text{ BkO}_2 \text{ on } 1.5 \mu\text{m-Ti foil}$

target-making
June 2009
First run:
from July 27 to October 23, 2009  Beam dose: $2.4 \times 10^{19}$
The discovery of a new chemical element with atomic number Z=117 is reported. The isotopes 293\(^{117}\) and 294\(^{117}\) were produced in fusion reactions between 48\(^{Ca}\) and 249\(^{Bk}\). Decay chains involving eleven new nuclei were identified by means of the Dubna Gas Filled Recoil Separator. The measured decay properties show a strong rise of stability for heavier isotopes with Z \(\geq 111\), validating the concept of the long sought island of enhanced stability for super-heavy nuclei.
Number of observed decay chains
Element 118     3
Element 116     26
Element 115     4
Element 114     43
Element 113     2
Element 112     8
Recognition of discovery of the elements 114 and 116

The IUPAC/IUPAP Joint Working Party (JWP) on the priority of claims to the discovery of new elements 113–116 and 118 has reviewed the relevant literature pertaining to several claims.

It was determined that the Dubna-Livermore collaborations share in the fulfillment of those criteria both for elements Z = 114 and 116.
Confirmation of the synthesis of the 117\textsuperscript{th} element
We are still far from the stability line

$^{277}_{112}$, $T_{1/2} \sim 0.2$ ms

$^{284}_{112}$, $T_{1/2} \sim 50$ s

$^{208}_{82}$ Pb + $^{70}_{28}$ Zn

$^{248}_{96}$ Cm + $^{48}_{20}$ Ca

Shift with 7 neutrons gives 6 orders of magnitude in half-life
Relatively long half-lives of isotopes of elements 104-116 produced in the reactions with $^{48}$Ca and chemical properties of SHE predicted theoretically permit new experiments aimed at:

- the chemical identification of SHE,
- study of their chemical properties,
- determination of masses of the SHE isotopes
Of-line chemical separation of $^{268}\text{Db}$

Dmitriev S N et al., *Mendeleev Commun.* 15 (2005) 1
Stoyer N J et al., *Proc. 9th Int. Conf. NN Collisions*, Brazil, 28 Aug.–1 Sep. 2006.

Experimental Setup

\[ v \left( A=48 \right) = 0.11 \text{ c} \]
\[ q = 16.5^+ \]

\[ v \left( A=288 \right) = 0.017 \text{ c} \]
\[ q = 6.2^+ \]
### Chemical Separation of $^{268}$Db - 2011

- **$^{48}$Ca + $^{243}$Am**

<table>
<thead>
<tr>
<th></th>
<th>2005</th>
<th>2011</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{243}$Am (mg cm$^{-2}$)</td>
<td>1.2</td>
<td>0.5</td>
</tr>
<tr>
<td>$\varepsilon_{\text{transm.}}$</td>
<td>0.9</td>
<td>0.4</td>
</tr>
<tr>
<td>$K(\text{Db/Ac})_{\text{sep.}}$</td>
<td>---</td>
<td>$10^4$-$10^5$</td>
</tr>
<tr>
<td>$K(\text{Db/Ac})_{\text{chem.}}$</td>
<td>$10^4$</td>
<td>$10^4$</td>
</tr>
</tbody>
</table>
Lanthanum fluoride radiochemical separation flow chart (4 h)

Copper foil (~400 mg)

Dissolution

Co-precipitation of group 4 and 5 elements (2 times)

Remove copper

Co-precipitation of group 4 elements

0.5 ml 1 M HF

Dissolve La(OH)₃

0.5 ml 1 M HCl

Removes traces of La, Cu

Cation Exchange Column
DOWEX50×8, 8×40 mm

1 M HF

Evaporation (1.5 ml)

Sat. Boric acid 2 M HNO₃

Group 4 Fraction
Dissolve LaF₃

Evaporation (1.5 ml)

Group 4 sample
65-70 % Hf

1 M HF
Elute Group 4 elements

3 M HF
Remove traces of La, Cu

Anion Exchange Column
DOWEX1×8, 6×12 mm

1.5% H₂O₂ / 1.5 M HNO₃
Elute Group 5 elements

Evaporation (0.5 ml)

Group 5 Fraction
0.5 M HCl / 0.5 M HF

Group 5 sample
70 -80 % Nb, 65-75% Ta

NH₄OH solution precipitate

Group 5 Fraction
0.5 M HCl / 0.5 M HF

Evaporation (0.5 ml)

Group 5 sample
70 -80 % Nb, 65-75% Ta

Group 4 Fraction
Dissolve LaF₃

Evaporation (1.5 ml)

Group 4 sample
65-70 % Hf

Anion Exchange Column
DOWEX1×8, 6×12 mm

1.5% H₂O₂ / 1.5 M HNO₃
Elute Group 5 elements

Evaporation (0.5 ml)
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</thead>
<tbody>
<tr>
<td>$N_{SF}$</td>
<td>3</td>
<td>18</td>
<td>21</td>
<td>15</td>
<td>5</td>
<td>8</td>
<td>28</td>
<td>49</td>
</tr>
<tr>
<td>$T_{1/2}$ (h)</td>
<td>$16^{+19}_{-6}$</td>
<td>$30^{+9}_{-6}$</td>
<td>$28^{+8}_{-5}$</td>
<td>$32^{+11}_{-7}$</td>
<td>$18^{+13}_{-6}$</td>
<td>$23^{+13}_{-6}$</td>
<td>$27^{+6}_{-4}$</td>
<td>$27.4^{+4.6}_{-3.4}$</td>
</tr>
<tr>
<td>$\sigma$ (pb)</td>
<td>8.5 $^{+6.4}_{-3.7}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>6.0 $^{+3.6}_{-2.4}$</td>
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</tbody>
</table>
Mendelev periodic table of the elements (2011)
GAS PHASE CHEMISTRY WITH ELEMENTS 112, 113 AND 114

- Are elements 112, 113 and 114 volatile metals?

- How do relativistic effects influence the chemistry of E112, 113 and of E114?
Relativistic Effects

\[ m = m_0 / \sqrt{1 - (v / c)^2} \]

\[ a_0 = 4\pi\varepsilon_0\hbar^2 / me^2 \]

\[ \Delta R_e(7s) = 20\% \]
How to experimentally determine a metallic character of a volatile element at a single atom level?

→ Determine interaction energy (adsorption enthalpy) with noble metals (e.g. Au)

→ If metallic: strong interaction (adsorption enthalpy) if non-metallic (noble gas like): weak interaction
Reaction: $^{242}\text{Pu}(^{48}\text{Ca},3n)^{287}114[0.5s] \rightarrow \alpha \rightarrow ^{283}112[3.6s]$

Compound Hg(Au) and 112(Au)
Reported at DGFRS:
Oganessian et al. 2004

Observed in Chemistry:

287\text{Ds}^{114}
- 1.1 s
- 10.04 MeV

283\text{Ds}^{112}
- 6.1 s
- 9.54 MeV

279\text{Ds}
- 0.29 s
- SF (>90%)
- 205 MeV

11.05.2006
- 2:40 (Moscow time)
- Det. 2 (T ~ -24°C)

25.05.2006
- 8:37 (Moscow time)
- Det. 7 (T ~ -5°C)

283\text{Ds}^{112}
- >3 s
- 9.45 MeV

287\text{Ds}^{114}
- >3 s
- 9.45 MeV

Detected at DGFRS:

25.05.2006
- 8:37 (Moscow time)
- Det. 7 (T ~ -5°C)

Detected in Chemistry:

11.05.2006
- 2:40 (Moscow time)
- Det. 2 (T ~ -24°C)

283\text{Ds}^{112}
- >3 s
- 9.53 MeV

279\text{Ds}
- 0.536 s
- SF
- 127+105 MeV

279\text{Ds}
- 0.592 s
- SF
- 108+123 MeV
Result from additional $^{48}$Ca + $^{242}$Pu experiments in 2007

Bombardement 21.3.- 17.4. 2007 with $3.1\times10^{18}$ $^{48}$Ca ions at $237\pm 3$ MeV
Element 112 is a noble metal – like Hg

- gas flow 0.86 l/min 
  He/Ar + Hg + Rn
- gas flow 0.89 l/min 
  He/Ar + Hg + Rn
- gas flow 1.5 l/min 
  He/Ar + Hg + Rn

- SF 231 MeV
- 0.53 s
- >140 MeV
- α 9.52 MeV
- 0.09 s
- 110 279 112 283
- 232 MeV SF
- >3 s
- 0.59 s
- 9.47 ± 0.12 MeV
- 110 279 112 283
- 231 MeV SF
- >3 s
- 0.77 s
- 9.35 ± 0.12 MeV
- 110 279 112 283
- 112+F2 MeV
- 0.77 s
- 85 ± 12 MeV SF
- 110 279 112 283

Element 112 is a noble metal – like Hg
$^{48}\text{Ca} + ^{244}\text{Pu}$

DGFRS

Chemistry

$^{284}\text{114}$

0.8 s

9.95 MeV

$^{288}\text{114}$

$T_{ads} = -84 \, ^\circ\text{C}$

$^{284}\text{112}$

101 ms

SF

$^{284}\text{112}$

$\tau: 109 \, \text{ms}$

SF

$^{288}\text{114}$

9.95 MeV
Result from the chemistry experiment with element 114

- Element 114 exhibits a very weak interaction with Au - pointing to a physisorptive interaction (similar to a noble gas).

- A quantitative description of this behaviour is lacking so far.
113 – 7s^2 7p_{1/2} – less reactive and more volatile than TL

- ΔH_{ads} (Au): Tl = 240 kJ/mol (S. König)
  113 = 158.6 (V. Pershina)

- ΔH_{ads} (inert surf) = 14 kJ/mol
Chemistry of the Element 113

$^{249}\text{Bk} + ^{48}\text{Ca}$

![Diagram of nuclear reactions and decay chains](image)
$^{48}\text{Ca} + ^{249}\text{Bk}$

Target:
- $^{249}\text{Bk}$ (0.5 mg·cm$^{-2}$)
- $^{\text{nat}}\text{Nd}$ (30 μg·cm$^{-2}$)

$^{48}\text{Ca}$

$E_{\text{mid. target}} = 249$ MeV
$I \sim 9$ eμA

Irradiation:
- 18.04.2010 – 31.05.2010;

target I - $3.5 \times 10^{18}$; target II - $5.6 \times 10^{18}$

$9.1 \times 10^{18}$
Target ($^{249}$Bk; $\approx$ 0.5 mg/cm$^2$)

SiO$_2$-

$800^\circ$C

(4$\pi$)

He/Ar (70/30)

2.5m

1 L/min

16 pairs

Au - 113

CHEMISTRY OF THE 113 ELEMENT
a) Cumulative alpha spectrum from 16 pairs of detectors
b) Cumulative spectrum of fission fragments from 16 pairs of detectors
c) Cumulative alpha spectrum from 4th pair of detectors
d) Cumulative spectrum of fission fragments from 4th pair of detectors

Counts / 5 keV
Counts / 0.05 MeV

Energy (keV)
Energy (MeV)

2 FF coincided Alphas
2 FF coincided
\[ E^* = 35 \text{ MeV} \]

1 event

TKE = 219(5) MeV
33.4 h

DGFRS
04 May 2010 10:05:46
Bk-target I

16 May 2010 02:29:54
Bk-target II
The deposition of $^{185}\text{Hg}$ compared to the deposition of $^{209}\text{At}$ in the isothermal detector array at $0^\circ\text{C}$ together with Monte-Carlo simulation of the depositions.
Chemistry of the Element 113

249Bk + 48Ca

281Rg

285115

289115

289115

284113

280Rg

278113

274113

270Db

272113

272113

268Db

TKE = 218(5) MeV
TKE = 219(5) MeV
TKE = 205(5) MeV

DGFRS
Experimental program of 2011-2012

- Synthesis of 115 element (243Am + 48Ca)
- Synthesis of 117 (119) element (249 Bk + 48Ca (50Ti))
- Mass number measurement of 112 - 114 elements

$^{242}\text{Pu}(^{48}\text{Ca},3n)^{287}114(0.5\text{s},\alpha) \rightarrow ^{283}112(4\text{ s})$

$^{243}\text{Am}(^{48}\text{Ca},3n) \rightarrow ^{288}115 (0.1 \text{ s}, \alpha) \rightarrow ^{284}113(0.5 \text{ s})$
STATUS of the MASS-SPECTROMETER

Mass-spectrometer MASHA at the beam line of the cyclotron U-400M
Mass-spectrometer “MASHA” status

Material of the catcher – flexible graphite
Operating temperature of hot catcher – 1500-1800°C
Delivery time of atoms from hot catcher to the ECR ion source ~ 1 s
Creation of the new experimental hall ($\approx 2600 \text{ m}^2$)
Creation of high current heavy ion accelerator
Development and creation of next generation set-ups
**NEW FLNR ACCELERATOR**

In order to improve efficiency of the experiments for the next 7 years it is necessary to obtain the accelerated ion beams with following parameters.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Energy</td>
<td>4÷8 MeV/n</td>
</tr>
<tr>
<td>Masses</td>
<td>10÷100</td>
</tr>
<tr>
<td>Intensity (up to 48Ca)</td>
<td>10 pμA</td>
</tr>
<tr>
<td>Beam emittance less 30 π mm·mrad</td>
<td></td>
</tr>
<tr>
<td>Efficiency of beam transfer</td>
<td>&gt;50%</td>
</tr>
<tr>
<td>ECR frequency</td>
<td>18÷28 GHz</td>
</tr>
</tbody>
</table>
Gas-Filled Separator

ADVANTAGES:

- presence of a gas – H₂, He;
- high energy and charge acceptances;
- additional target cooling through convection
- low number of elements;
- no high voltage.
### General ion-optical parameters of “MASHA” with gas catcher

<table>
<thead>
<tr>
<th>Parameters</th>
<th>ECR ion source</th>
<th>Gas catcher</th>
</tr>
</thead>
<tbody>
<tr>
<td>Range of energy variation, kV</td>
<td>15-40</td>
<td>15-40</td>
</tr>
<tr>
<td>Range of Bp variation, Tm</td>
<td>0.08-0.5</td>
<td>0.08-0.5</td>
</tr>
<tr>
<td>Mass acceptance, %</td>
<td>2.8</td>
<td>2.8</td>
</tr>
<tr>
<td>Angular spread, mrad</td>
<td>14</td>
<td>5</td>
</tr>
<tr>
<td>Diameter the ion source exit hole, mm</td>
<td><strong>5.0</strong></td>
<td><strong>1.0</strong></td>
</tr>
<tr>
<td>Horizontal magnification at F1/F2</td>
<td><strong>0.39/0.68</strong></td>
<td><strong>0.24/0.90</strong></td>
</tr>
<tr>
<td>Vertical magnification at F1/F2</td>
<td><strong>2.4/3.13</strong></td>
<td><strong>1.5/1.25</strong></td>
</tr>
<tr>
<td>Linear mass resolution at F1</td>
<td>75</td>
<td>420</td>
</tr>
<tr>
<td>Mass resolution at F2</td>
<td><strong>1150</strong></td>
<td><strong>5700</strong></td>
</tr>
</tbody>
</table>
FROM ISOL TECHNIQUE TO GAS CATCHER

Gas catcher for heaviest nuclei research

Geometry
- Entrance foil – Ti (d=2 mm, Ø=100 mm)
- Diameter of the gas cell – 160 mm
- Length of the gas cell – 210 mm
- Exit nozzle diameter – 1,2 mm

Main parameters
- Operating gas – He purity ~1 ppb.
- Pressure into gas cell – 100-200 mbar.
- Gas flow from the nozzle – 90-120 mbar·l/s
- Extraction time ~ 10 ms.
- Efficiency 10-40%.
- Beam emittance ~3.0 π·mm·mrad
Первый этаж на отметках 0,5+1,0
DRIBs-III: New experimental building
Thank you

FLNR, JINR (Dubna)
ORNL (Oak-Ridge, USA)
LLNL (Livermore, USA)
RIAR (Dimitrovgrad, Russia)
Vanderbilt University (Nashville, USA)
## New resources and research opportunities

<table>
<thead>
<tr>
<th>Beam</th>
<th>E/A (MeV)</th>
<th>Intensity (ppps)</th>
<th>E/A (MeV)</th>
<th>Intensity (ppps)</th>
<th>E/A (MeV)</th>
<th>Intensity (ppps)</th>
<th>E/A (MeV)</th>
<th>Intensity (ppps)</th>
<th>Physics</th>
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<tr>
<td>light RIB</td>
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</tr>
<tr>
<td>6He</td>
<td>35</td>
<td>4×10^{13}</td>
<td>35</td>
<td>6×10^{12}</td>
<td>35</td>
<td>2.5×10^{12}</td>
<td></td>
<td></td>
<td>structure of light exotic nuclei, reactions, sub-barrier fusion, astrophysics</td>
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<tr>
<td>8He</td>
<td>33</td>
<td>8×10^{12}</td>
<td>33</td>
<td>2×10^{12}</td>
<td>33</td>
<td>6×10^{11}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>24Ne</td>
<td>40</td>
<td>7×10^{11}</td>
<td>40</td>
<td>4×10^{11}</td>
<td>35</td>
<td>6×10^{11}</td>
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<td></td>
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<tr>
<td>6&lt;A≤40</td>
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<td>6&lt;A≤40</td>
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<td>6&lt;A≤40</td>
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<td>7Li</td>
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<td>40Ar</td>
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<td>2.5×10^{11}</td>
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<td>A~60</td>
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<td>48Ca</td>
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<td>4×10^{12}</td>
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<td>3×10^{11}</td>
<td>5</td>
<td>6×10^{11}</td>
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<td>SIE with Z&gt;118 (σ=0.1 pb) spectroscopy of SHE, fission-fission, quasi-fission</td>
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<tr>
<td>54Cr</td>
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<td>58Fe</td>
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<td>A~150</td>
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<td>124Sn</td>
<td>5</td>
<td>3×10^{11}</td>
<td>5</td>
<td>2×10^{11}</td>
<td>5</td>
<td>2×10^{11}</td>
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<td></td>
<td>deep inelastic scattering, multi-nucleon transfer, new neutron rich nuclei, shell effects</td>
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<td>A~240</td>
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<tr>
<td>238U</td>
<td>7</td>
<td>3×10^{10}</td>
<td>7</td>
<td>2×10^{10}</td>
<td>7</td>
<td>10^{11}</td>
<td></td>
<td></td>
<td>neutron-rich SHE, new heavy isotopes, ternary fission, super strong electric fields, e⁺-e⁻</td>
</tr>
</tbody>
</table>