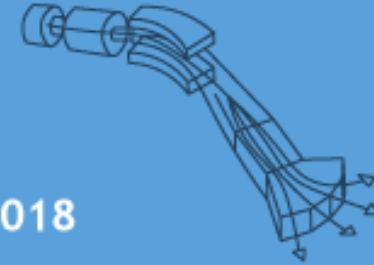


# EMIS XVIII



CERN GENEVA / SWITZERLAND / 16 - 21 SEPTEMBER 2018



## Molecular beams in the ISOL process

Jochen Ballof<sup>1,2</sup>

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<sup>5</sup> Institut für Kernphysik, Technische Universität Darmstadt, Germany

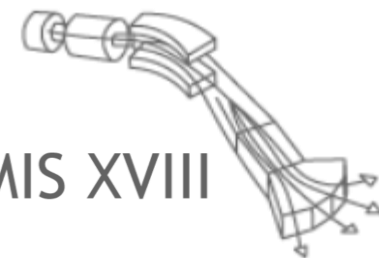
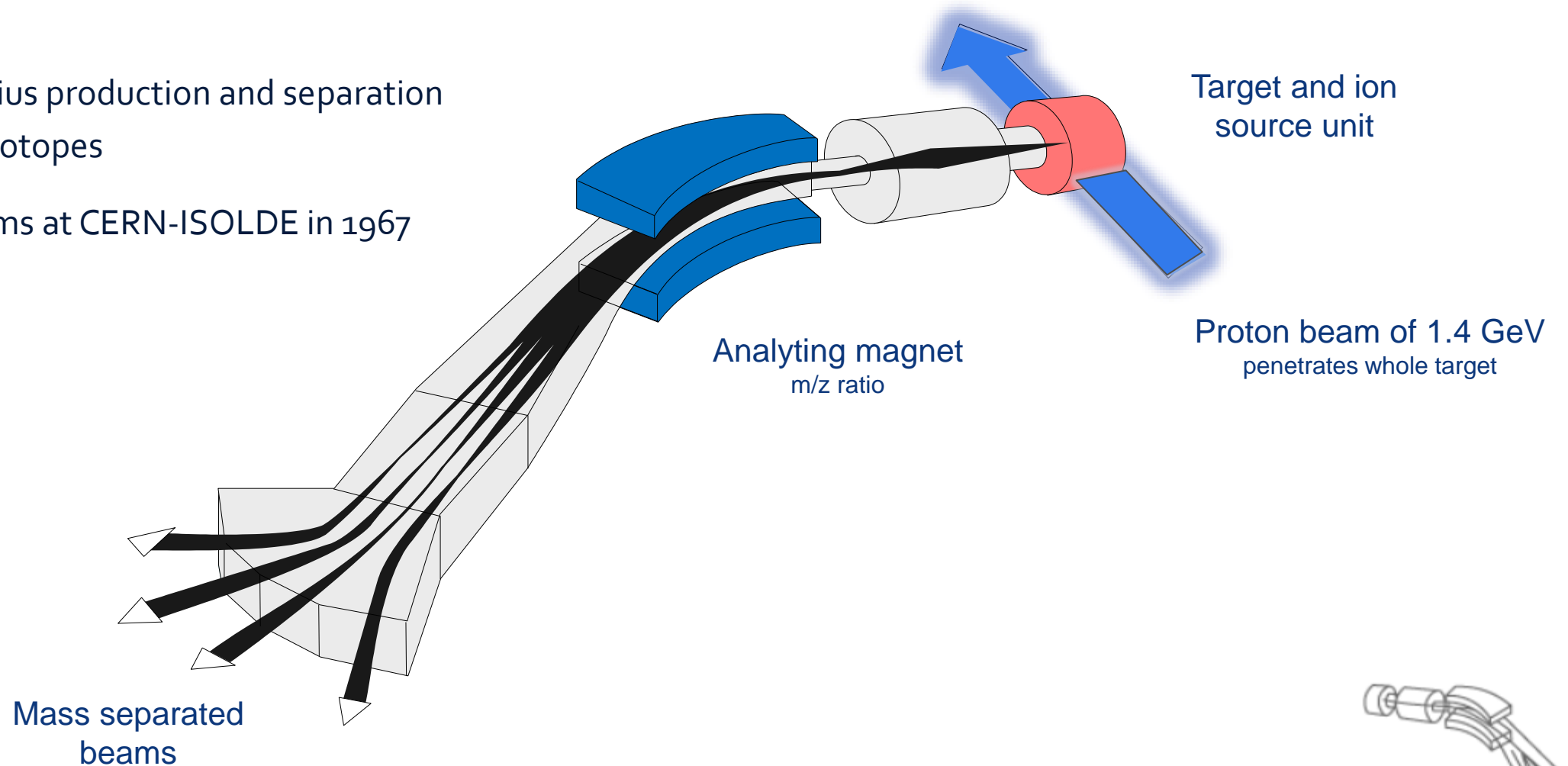


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# The ISOL process

Isotope Separation On Line

- Simultaneous production and separation of radioisotopes
- First beams at CERN-ISOLDE in 1967



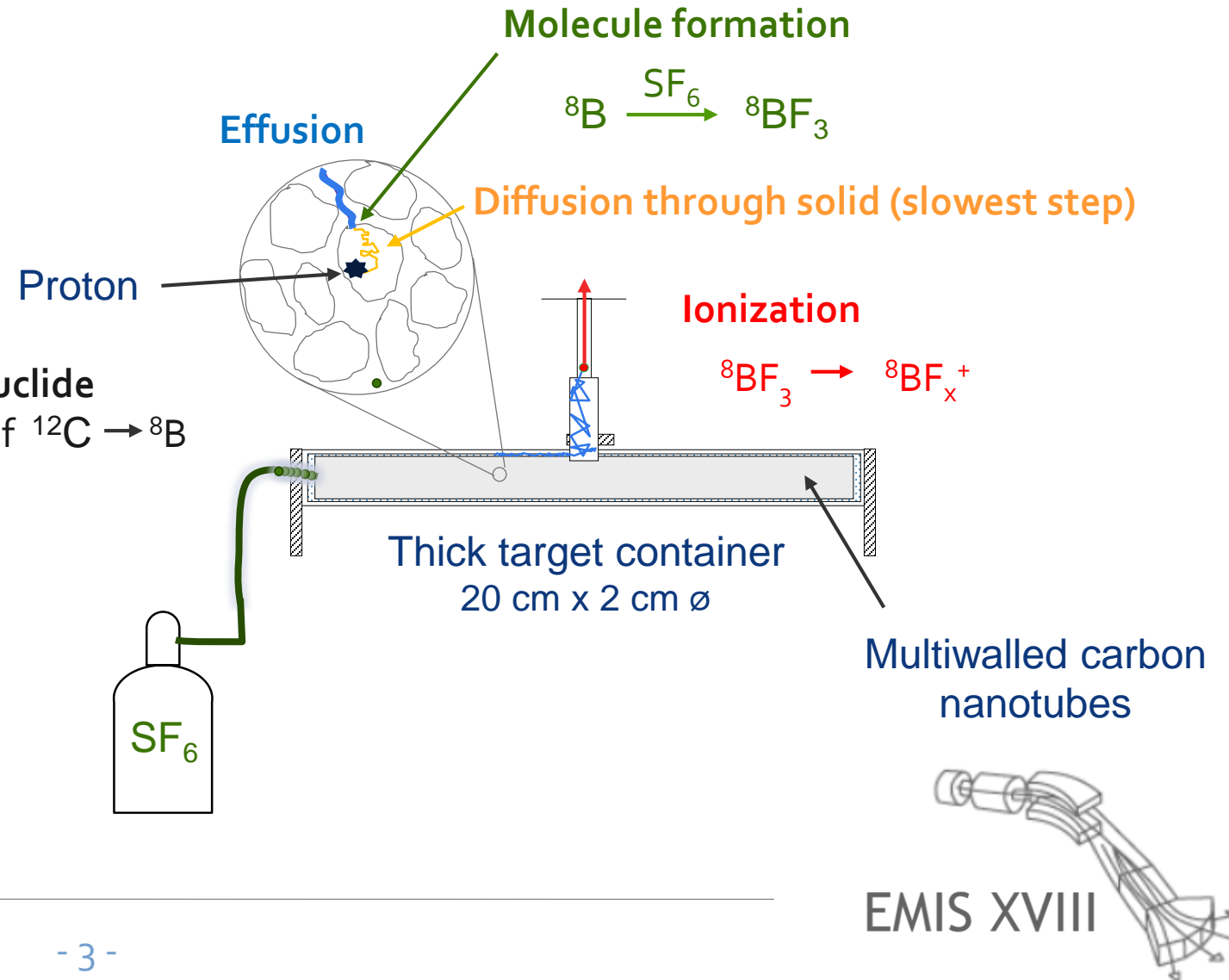
# The ISOL process

## Molecular beam extraction taking the example of ${}^8\text{BF}_2$

- Boron is refractory and reactive
- Can't be extracted as atom -> more volatile molecule needed

Production of nuclide  
Fragmentation of  ${}^{12}\text{C} \rightarrow {}^8\text{B}$

$$\text{Yield} = N_0 \times \epsilon_{\text{release}} (T_{1/2}) \times \epsilon_{\text{form}} \times \epsilon_{\text{ion}} \times \epsilon_{\text{other}}$$



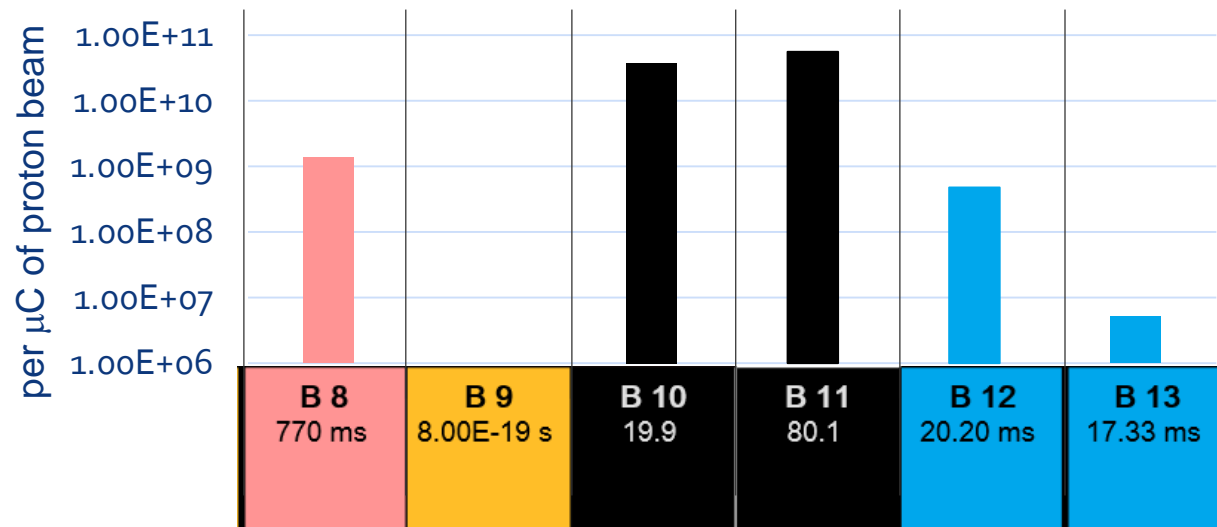
# Boron beams

Boron isotopes

**in-target** production from multiwalled carbon nanotubes by 1.4 GeV protons

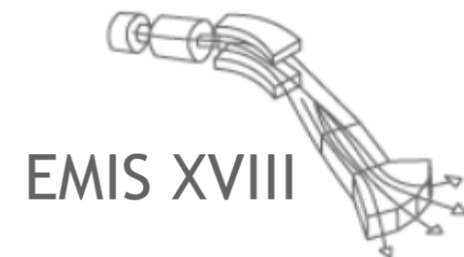
FLUKA / ABRABLA

$$\text{Yield} = N_0 \times \epsilon_{\text{release}} (T_{1/2}) \times \epsilon_{\text{form}} \times \epsilon_{\text{ion}} \times \epsilon_{\text{other}}$$



ISOLDE:  
typical. max. intensity:  
 $2 \mu\text{A}$

Simulations:  
Joao-Pedro Ramos, Christoph Seiffert

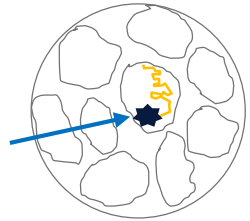


# Boron beams

Release Efficiency: Fraction of isotopes not decayed before release

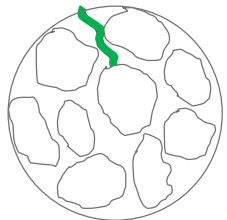
$$\text{Yield} = N_0 \times \epsilon_{\text{release}}(T_{1/2}) \times \epsilon_{\text{form}} \times \epsilon_{\text{ion}} \times \epsilon_{\text{other}}$$

## 1. Diffusion in the solid



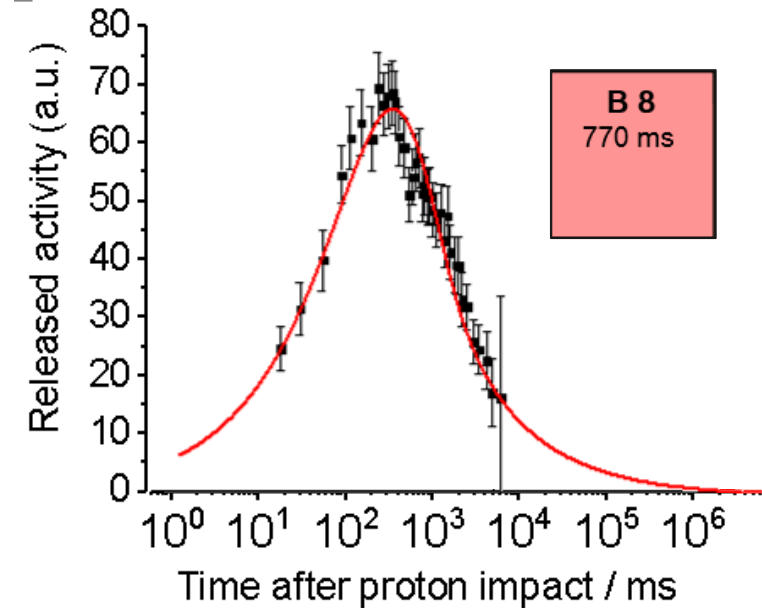
- Slow process
  - Was investigated in offline experiments
  - Long heat treatment times necessary ~ 30 min
  - Simple model: Fick's 2<sup>nd</sup> law
- ➔ **Characteristic diffusion time**  $\tau_D = 862 \text{ s}$

## 2. Effusion through open space



- Fast process
  - Best investigated online
- ➔ **Effusion time constant**  $t_{\text{eff}} = 339 \text{ ms}$

Online release curve fitted with release model measured on <sup>8</sup>B and corrected for decay



Model for release of a stable isotope:

$$p(t) = \frac{1}{A_K} \sum_{m=1}^{\infty} \frac{e^{-c_m t / \tau_D} - e^{-t / t_{\text{eff}}}}{\tau_D - c_m t_{\text{eff}}}$$

with  $c_m = (j_{0,m} / \pi)^2$ ,  $j_{0,m}$   $m^{\text{th}}$  positive root of Bessel function of order 0,  $A_K$  normalization constant

Offline mobility studies: Christoph Seiffert

# Boron beams

## Release Efficiency

$$\text{Yield} = N_0 \times \epsilon_{\text{release}}(T_{1/2}) \times \epsilon_{\text{form}} \times \epsilon_{\text{ion}} \times \epsilon_{\text{other}}$$

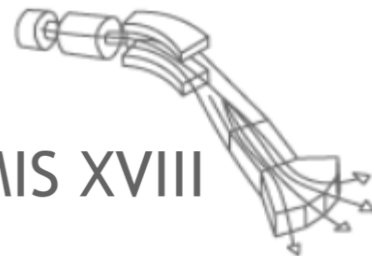
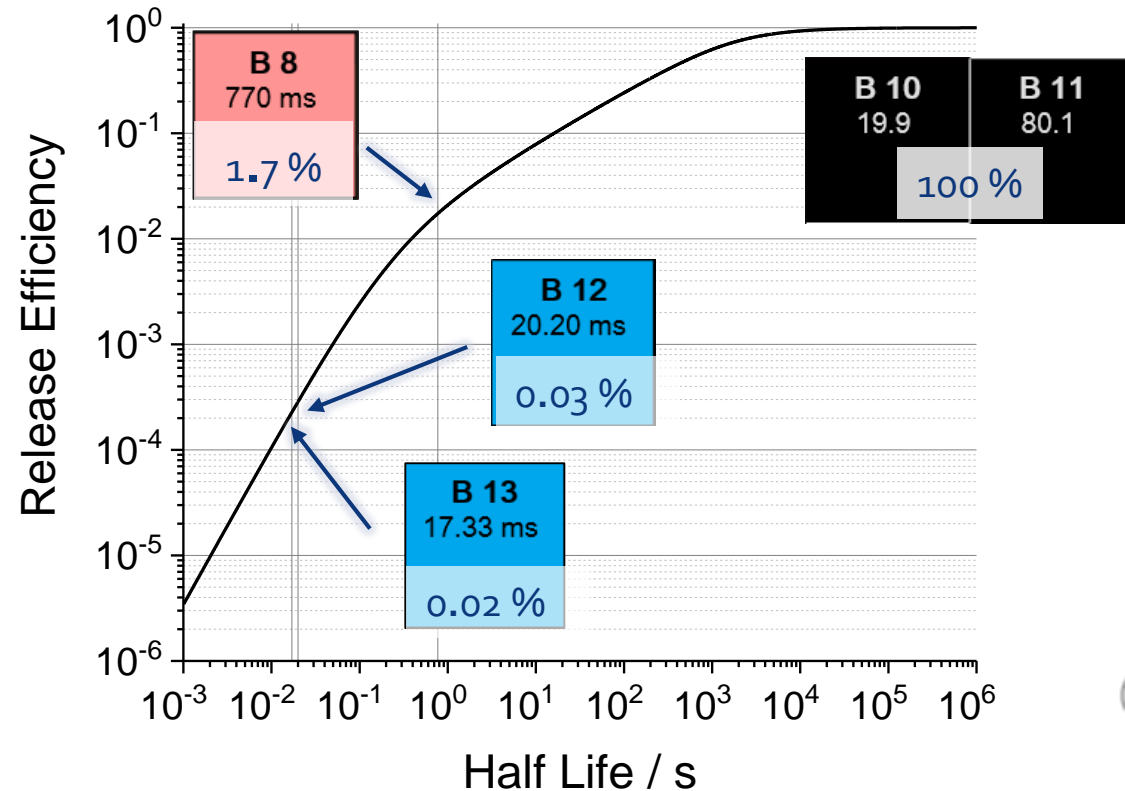
Release efficiency can be obtained from release curve:

$$\epsilon_{\text{release}}(T_{1/2}) = \int_0^{\infty} p(t) e^{-\ln(2) t/T_{1/2}} dt$$

Function holds for all isotopes of the same chemical elements.



Allows prediction of yields for more exotic isotopes  $^{12}\text{B}$ ,  $^{13}\text{B}$



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# Boron beams

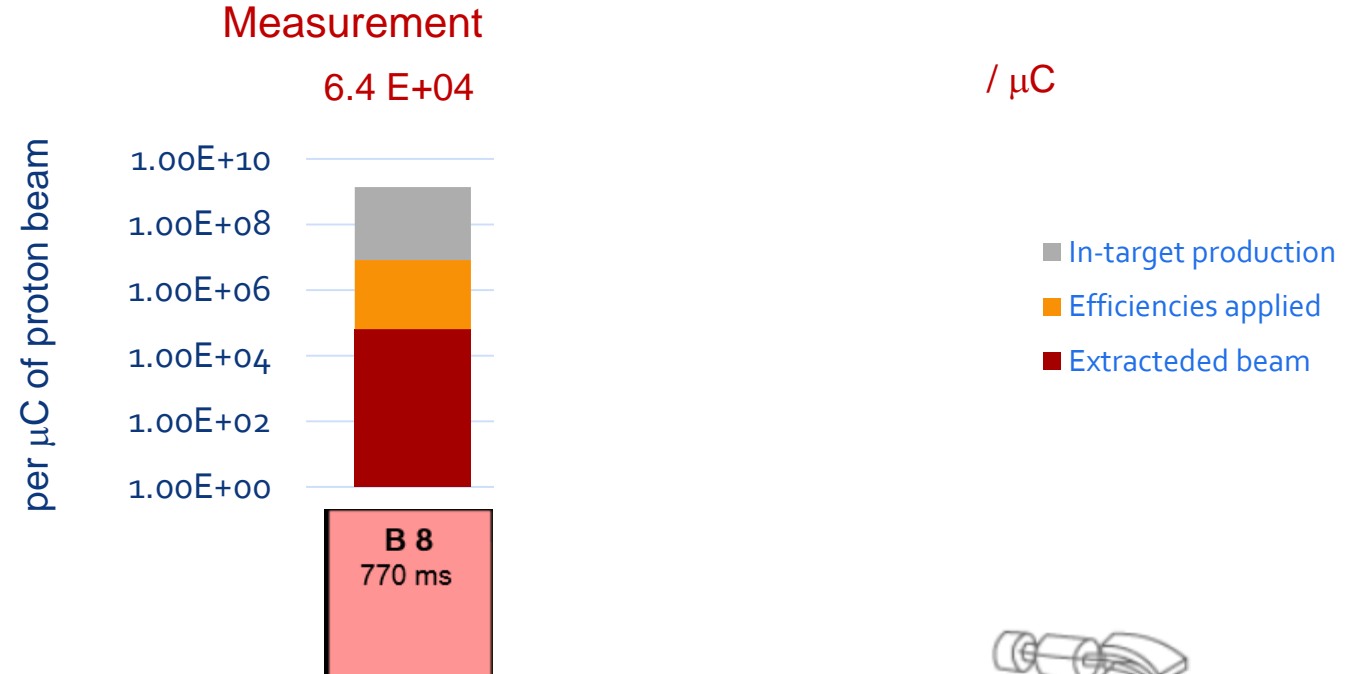
Putting it all together

$$\text{Yield} = N_0 \times \varepsilon_{\text{release}} (T_{1/2}) \times \varepsilon_{\text{form}} \times \varepsilon_{\text{ion}} \times \varepsilon_{\text{other}}$$

- Other efficiencies are typically assumed to be independent of the half life
- Can be calculated, if ...
  - o the **yield** of one isotope was measured
  - o  $N_0$  is known from simulations
  - o  $\varepsilon_{\text{release}} (T_{1/2})$  was calculated
- Or measured offline

Measured yield on  ${}^8\text{BF}_2$ :  $6.4 \times 10^4 / \mu\text{C}$

$$\rightarrow \varepsilon_{\text{form}} \times \varepsilon_{\text{ion}} \times \varepsilon_{\text{other}} = 0.6 \%$$



*J. Ballof, C. Seiffert et. al, submitted to EPJA (2018)*

# Boron beams

Testing the

Yield =  $N_0$

M

per  $\mu\text{C}$  of proton beam

$1.00\text{E}+10$   
 $1.00\text{E}+09$   
 $1.00\text{E}+08$   
 $1.00\text{E}+07$   
 $1.00\text{E}+06$   
 $1.00\text{E}+05$   
 $1.00\text{E}+04$   
 $1.00\text{E}+03$   
 $1.00\text{E}+02$   
 $1.00\text{E}+01$

## The ISOLDE Yield Database

### A new tool to predict beam intensities

J. Ballof<sup>1,2</sup>, J. P. Ramos<sup>2</sup>, A. Molander<sup>2,3</sup>, K. Johnston<sup>2</sup>, S. Rothe<sup>2</sup>, T. Stora<sup>2</sup>

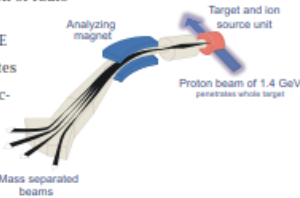
1) Johannes Gutenberg - Universität Mainz, Saarstr. 21, 55122 Mainz, Germany    2) CERN, ISOLDE, 1211 Geneva 23, Switzerland    3) University of Helsinki, 00014 University of Helsinki, Finland

Developed more than 10 years ago, the ISOLDE yield database [1,2] serves as a valuable source for experiment planning.

With the increasing demand for more and more exotic beams, needs arise to extend the functionality of the database and website not only to provide information about yields determined experimentally, but also to predict yields of isotopes, which can only be measured with sophisticated setups.

#### Isotope Separation OnLine

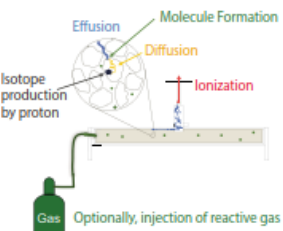
- Simultaneous production and separation of radio-isotopes
- First applied in 1951 at CERN -> ISOLDE
- Thick targets allow high production rates
- Variety of target materials and ion-sources available
- Supplied by the Proton Synchrotron Booster (PSB) with 1.4 GeV protons
- Proton intensities up to 2  $\mu\text{A}$



#### Extraction Process

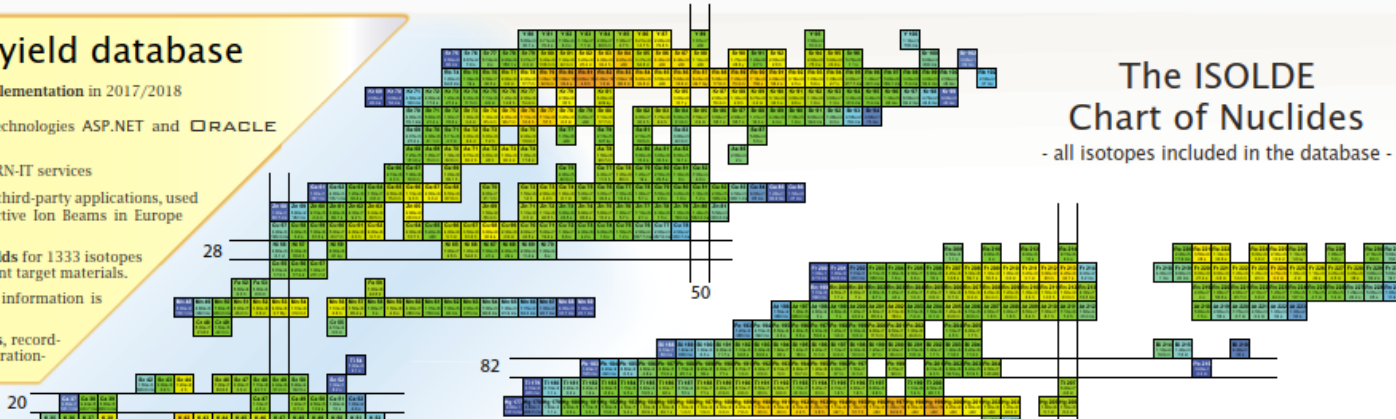
1. Isotope production in grain of target material ( $N_0$ )
2. Diffusion (typically slow) to the surface of the grain
3. In some cases: Formation of a volatile molecule
4. Effusion from the target container to the ion source
5. Ionization

**Yield:**  
 $Y = N_0 \epsilon_{\text{Release}} \epsilon_{\text{Formation}} \epsilon_{\text{Ionization}} \epsilon_{\text{Other}}$



#### The ISOLDE yield database

- Complete Redesign and -implementation in 2017/2018
- Using state-of-the-art web technologies ASP.NET and ORACLE databases
- Hosted and supported by CERN-IT services
- Open XML Web-interface for third-party applications, used e.g. by the Chart of Radioactive Ion Beams in Europe (CRIBE) project [3]
- Contains 2445 evaluated yields for 1333 isotopes of 74 elements and 55 different target materials.
- The Release time structure information is available for 427 yields.
- Newly added technical yields, recorded continuously for each operational target.
- Results of high-statistics Monte-Carlo sim-

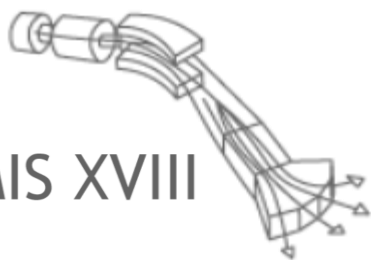


#### The ISOLDE Chart of Nuclides

- all isotopes included in the database -


Poster tomorrow

Preliminary  
 ISOLDE (S) C  
 García Borge, (GISOL)






# More molecular beams - Posters





**Prospects for the production of  $^{100}\text{Sn}$  ISOL beams at HIE-ISOLDE**  
F. Boix Pamies, R. Catherall, S. Malbrunot, G. Neyens, J.P. Ramos, S. Rothe, T. Stora  
CERN, CH-1211 Geneva 23, Switzerland



Context


→ Extraction as  $\text{SnCl}_x$



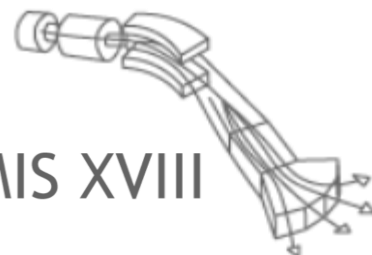

**Infrastructure for the Production and Development of Targ**  
**CERN-ISOLDE**

Sebastian Rothe<sup>1</sup>, Jochen Ballof<sup>1,3</sup>, Ermanno Barbero<sup>1</sup>, Ferran Boix Pamies<sup>1</sup>, Richard Catherall<sup>1</sup>, Katerina Chrysalidis<sup>1,2</sup>, Bernard Crepieux<sup>1</sup>, Kevin Develle<sup>1</sup>, Paul Harwood<sup>1</sup>, Reinhard Heinke<sup>2</sup>, David Leimbach<sup>1,2</sup>, Annie Ringvall-Moberg<sup>1,4</sup>, Bruce Marsh<sup>1</sup>, Michael Owen<sup>1</sup>, João Pedro Ramos<sup>1</sup>, Ralf Erik Rossel<sup>1</sup>, Thierry Stora<sup>1</sup>, Andres Suarez<sup>1</sup>, Nhat-Tan Vuong<sup>1,5</sup>, Shane Wilkins<sup>1</sup>

<sup>1</sup>CERN, Geneva, Switzerland  
<sup>2</sup>Institut für Physik, Johannes Gutenberg-Universität, Mainz, Germany  
<sup>3</sup>Institut für Kernchemie, Johannes Gutenberg-Universität, Mainz, Germany  
<sup>4</sup>Department of Physics, Gothenburg University, Gothenburg, Sweden  
<sup>5</sup>Doctoral School of Chemistry and Chemical Engineering, EPFL, Lausanne, Switzerland



EN Engineering Department



# Refractory Metal Carbonyl beams

Un(available) beams at ISOLDE  
and selected sidebands

1																	2
H																	He
3	4															10	
Li	Be															Ne	
11	12															18	
Na	Mg															Ar	
19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
55	56	71	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86
Cs	Ba	La...	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn

➔ potentially  
9 new beams!

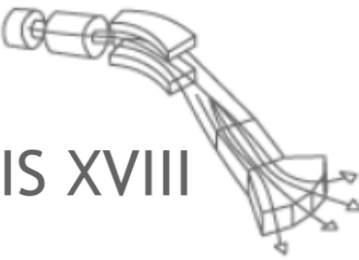
BF<sub>x</sub>

GeS  
SnS

SeCO

Available Beams	Unavailable Beams
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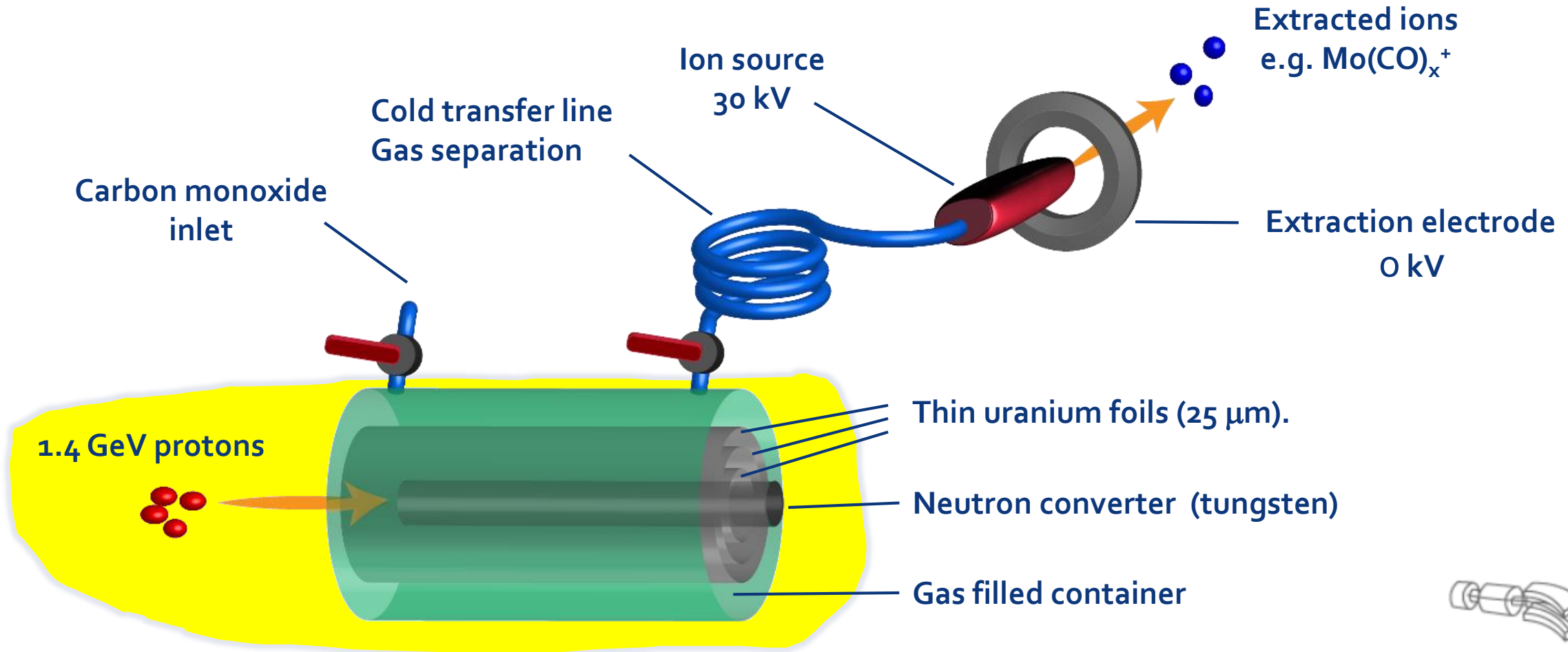
Volatile Compounds:    Fluoride    Carboxide    Sulfide    Carbonyl



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# Refractory Metal Carbonyl beams

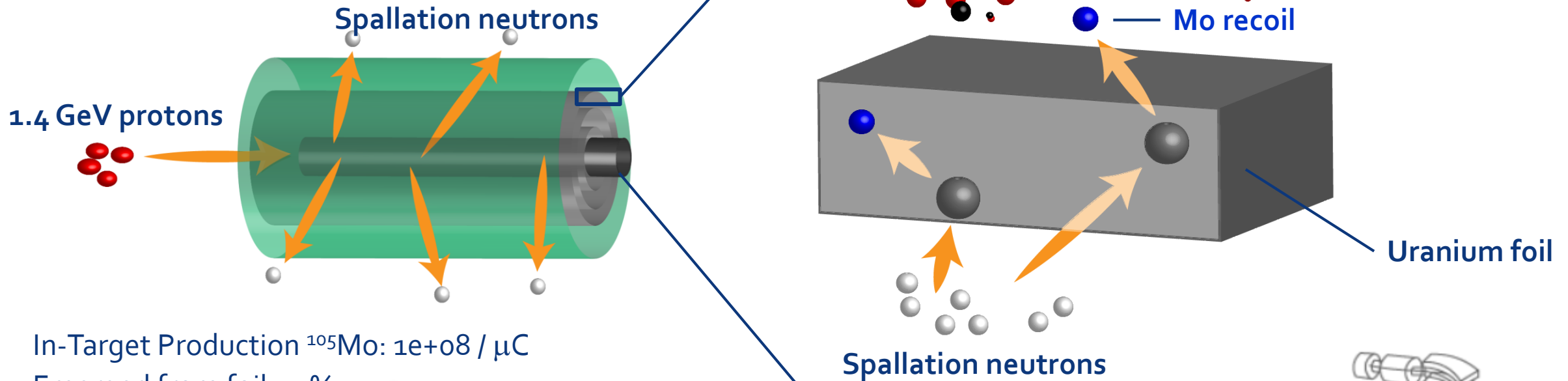
## The target concept



# Refractory Metal Carbonyl beams

## The target concept

- neutron converter to avoid beam on gas container
- Avoid slow diffusion through solids, use **recoil effect**
- *in-situ* volatilization: thermalizing recoils in CO gas



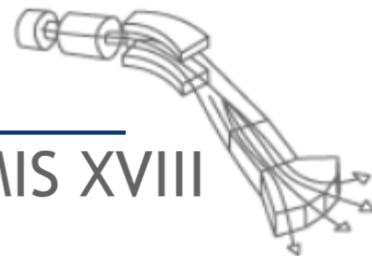
In-Target Production <sup>105</sup>Mo: 1e+08 / μC

Emerged from foil: 11%

Stopped in the gas: 51%



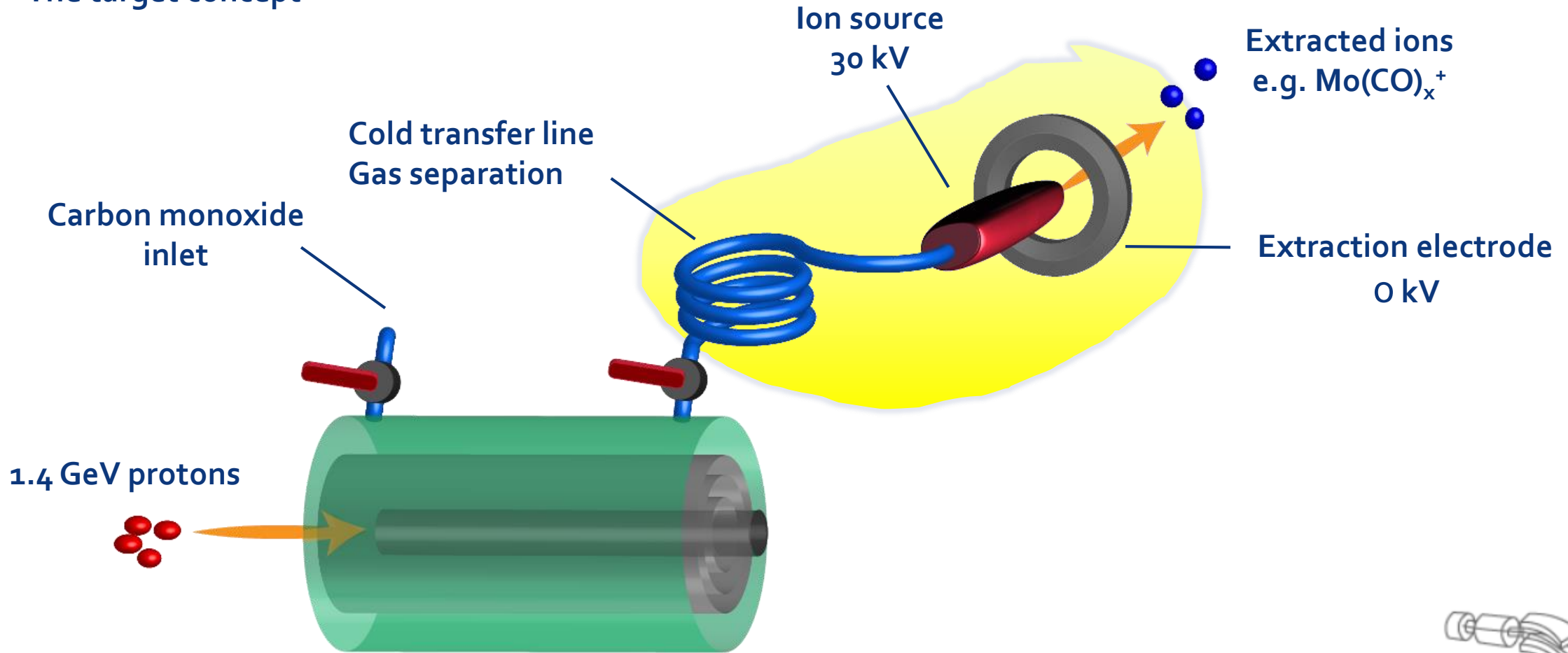
$$\epsilon_{\text{release}} = 5\%$$



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# Refractory Metal Carbonyl beams

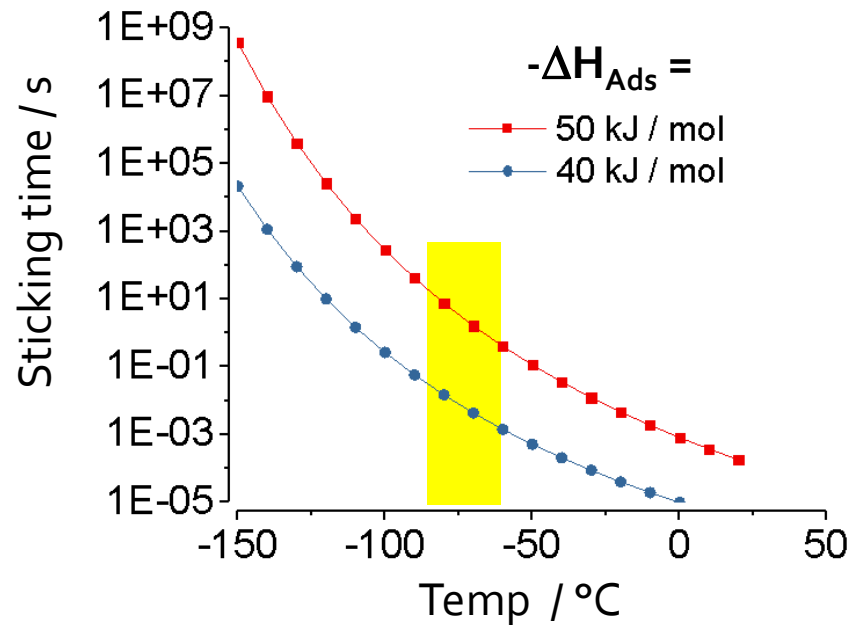
The target concept



# Refractory Metal Carbonyl beams

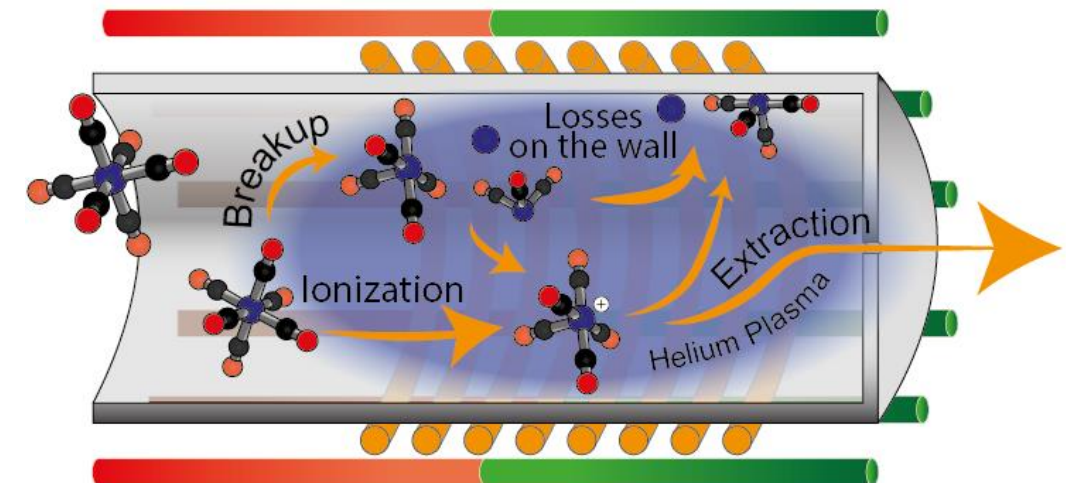
## Gas separation

- $\text{Mo(CO)}_6$  retained on cooling trap while gas is pumped
- Adsorption enthalpies on quartz surfaces known
  - > *Even et. al, Radiochim. Acta 2014*
- Similar set up for gas separation tested
  - > *Katagiri et. al, Rev. Sci. Instr. 2014*



## Ionization

- Ion sources should be cold,  $\text{Mo(CO)}_6$  decomposes easily
- Ionization tested with RF-Sources (HELICON, COMIC) and FEBIAD type sources
- Ionization feasible, but efficiency (so far) too low for radioactive beams.



# Summary

## First boron beams from thick ISOL targets

<b>B 8</b> 770 ms	<b>B 12</b> 20.20 ms	<b>B 13</b> 17.33 ms
----------------------	-------------------------	-------------------------

- Measurement of yield and release properties for  ${}^8\text{BF}_2$  ( $4.6 \text{ e}+04 / \mu\text{C}$ )
- Prediction of the extraction of exotic  ${}^{12}\text{B}$  and  ${}^{13}\text{B}$

## History

- 2015: First successful prototype
- 2016: HIE-ISOLDE Tests
- 2017: First low energy beam to users
- 2018: First accelerated beam to users

## Concept for the extraction of the most refractory metals

<b>Mo</b>	<b>Tc</b>	<b>Ru</b>	<b>Rh</b>
<b>W</b>	<b>Re</b>	<b>Os</b>	<b>Ir</b>

- Extraction from target foils by fission recoil effect
- *in-situ* volatilization by carbonyl formation
- cryogenic gas separation
- Ionization in ECR or Laser-based ion source

## Outlook

- Irradiation experiments planned to study carbonyl formation and survival at ISOLDE
- Investigation of ECR ion-sources

# Thanks for your attention!

Special thanks to

Ermanno Barbero, Vincent Barozier, Ana-Paula Bernardes, Ferran Boix Pamies, Richard Catherall, James Cruikshank, Melanie Delonca, Katerina Chrysalidis, Maria Garcia Borge, Paul Harwood, Karl Johnston, David Leimbach, Razvan Lica, Miguel Madurga Flores, Bruce Marsh, Tania Mendonca, Michael Owen, Yisel Martinez Palenzuela, Julien Riegert, Andres Suarez, Olof Tengblad, Miranda Van Stenis, Andree Welker and Shane Wilkins



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