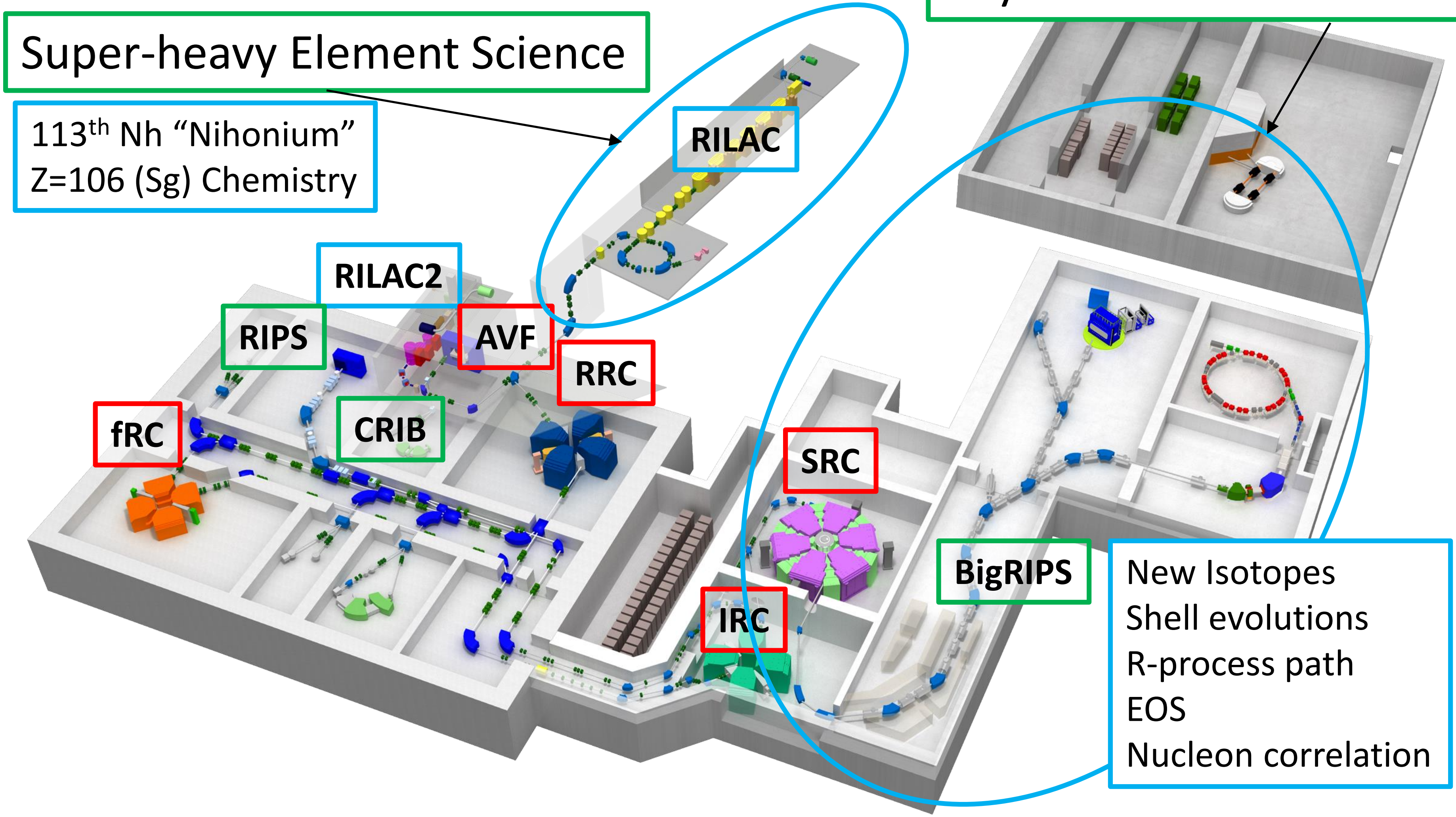
Long-lived Fission Product Transmutation

H. Sakurai
RIKEN Nishina Center

RI Beam Factory

5 cyclotrons + 2 linacs

3 inflight separators



Super-heavy Element Science

113th Nh “Nihonium”
Z=106 (Sg) Chemistry

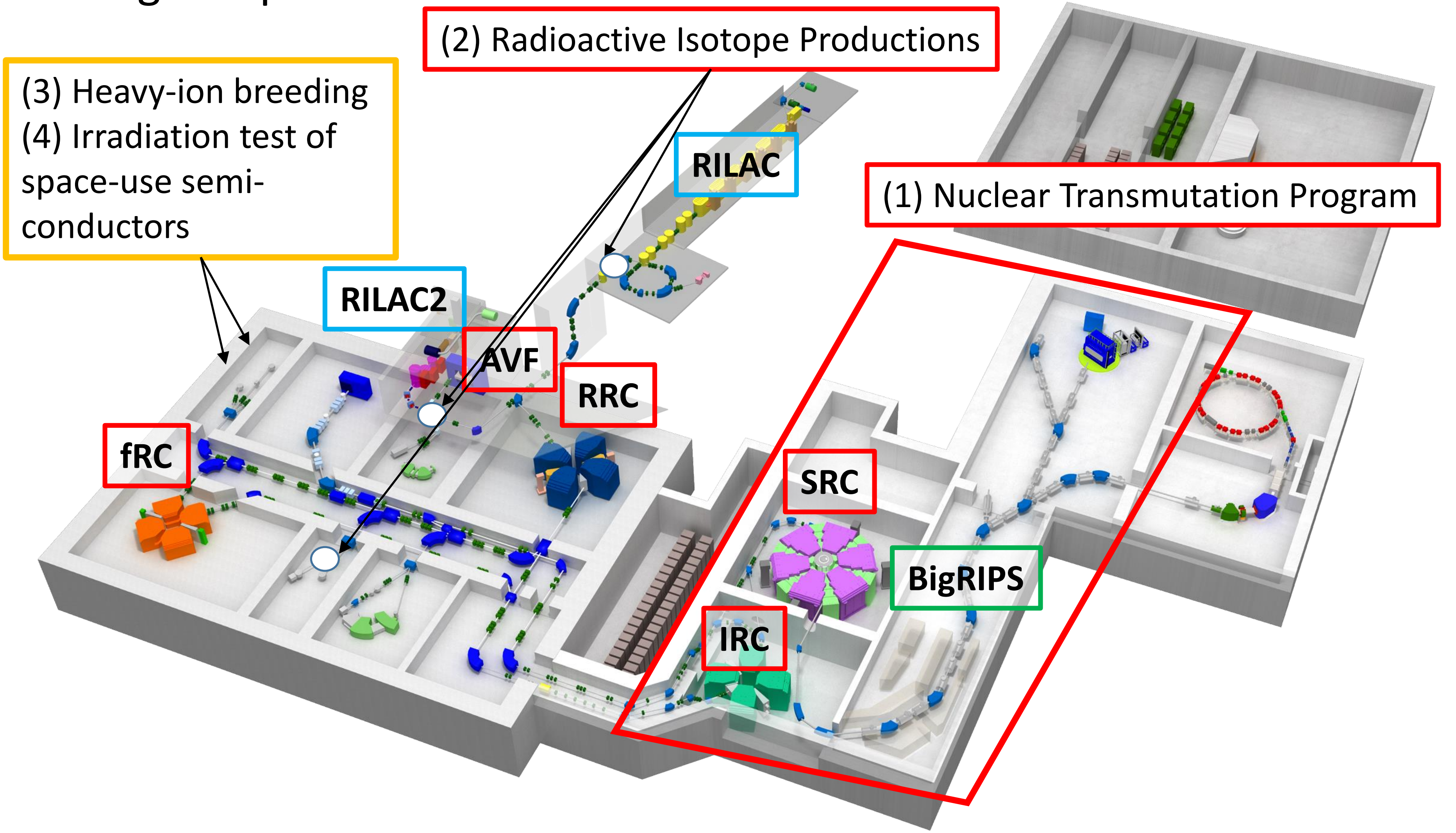
Physics with Exotic Nuclei

New Isotopes
Shell evolutions
R-process path
EOS
Nucleon correlation

Application Programs at RIBF

5 cyclotrons + 2 linacs

3 inflight separators



(2) Radioactive Isotope Productions

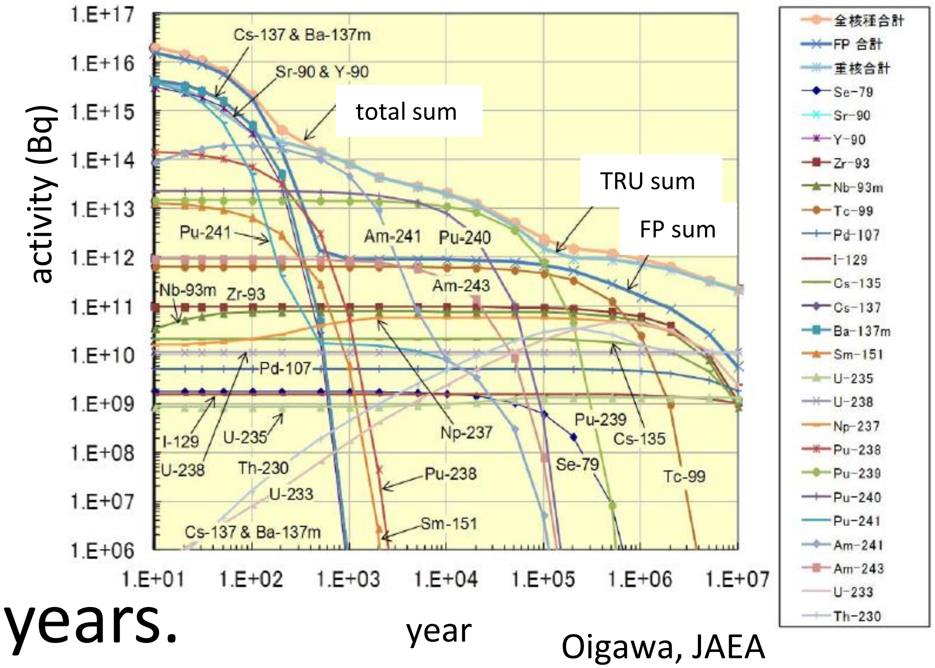
(3) Heavy-ion breeding

(4) Irradiation test of space-use semi-conductors

(1) Nuclear Transmutation Program

High-Level Nuclear Waste Problems

Nuclear Waste has long-lived fission products (FP) and minor actinoid (MA).



Oigawa, JAEA

In case of geological disposal, nuclear waste has a potential risk over a few ten thousands years.

Site of geological disposal is hardly selected.

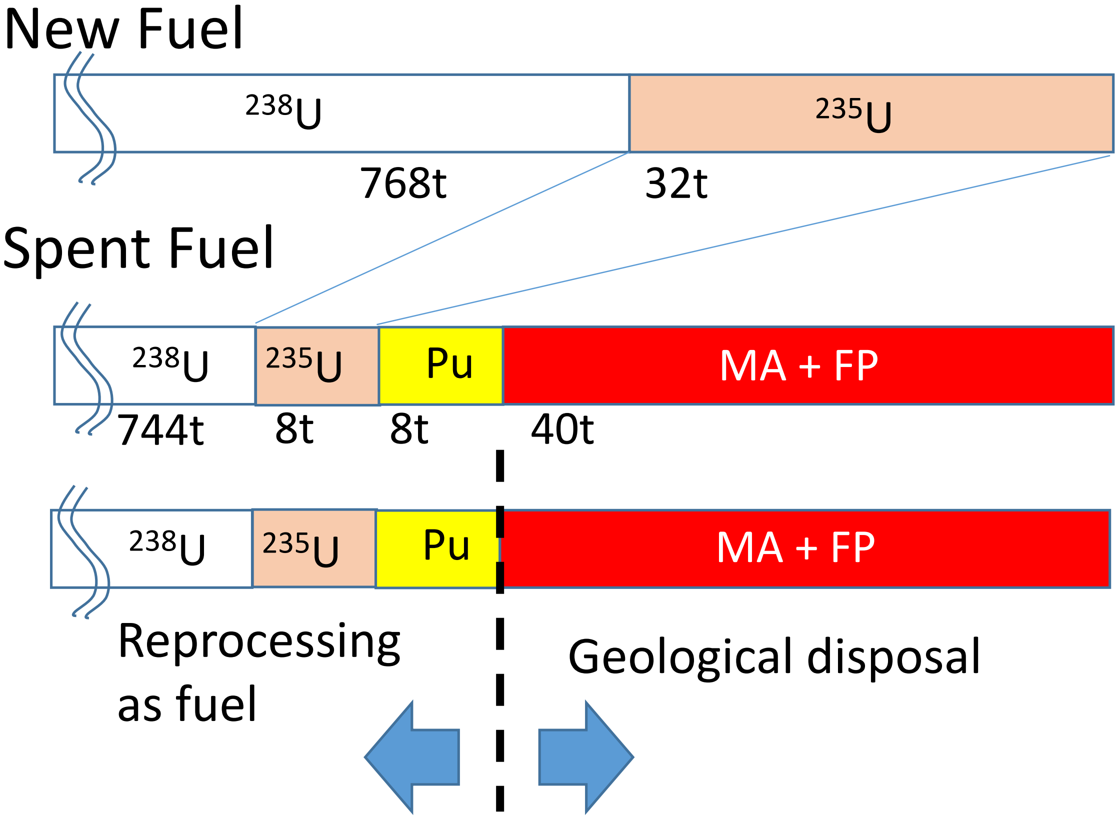
It is hard to guess how structures of ground layers will be changed for the coming ten thousands years.

Nuclear waste could be efficiently transmuted into harmless materials?

No leaving waste for next generation

Geological Disposal and/or Reprocessing

In Japan, before Fukushima Incident 2011,
~ 800t U / year (~75% operation of 50 LWR)



New Fuel

^{238}U 768t

^{235}U 32t

Spent Fuel

^{238}U 744t

^{235}U 8t

Pu 8t

MA + FP 40t

^{238}U ^{235}U Pu | MA + FP

Reprocessing
as fuel

Geological disposal

<u>Direct Disposal</u>

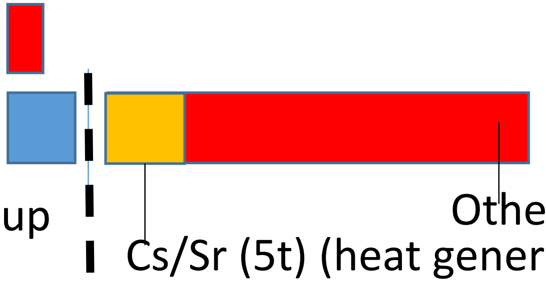
USA, Canada,
Finland, Sweden

<u>Reprocessing + Disposal</u>

France, Japan …

Further Partitioning and Reprocessing

R&D efforts to minimize risk of radioactive materials in future



MA (Np, Am, Cm) 1t → ADS, FBR

FP 39t

Platinum Group (4t) Pt, Pd, …
> re-used

Cs/Sr (5t) (heat generator)

Other FP (30t)

→ FBR

How about accelerator system to reduce radioactivity of FP?

A variety of reactions

Nuclear Reactions
Beam species, energy
Target material & system…..

⟺

Facility building cost
operation cost …

Lack of nuclear reaction data for FP (so far, n-capture only)

A challenge at RIKEN

First targets are Cs-137 and Sr-90, to study spallation reaction induced by proton and deuteron

Cs-137 and Sr-90 have a large weight fraction of FP.

Half-life is about 30 years.

At the present policy, “cooling” time needs about 300 years.

The thermal neutron capture cross sections are small.

0.27 b for Cs-137, 0.01 b for Sr-90

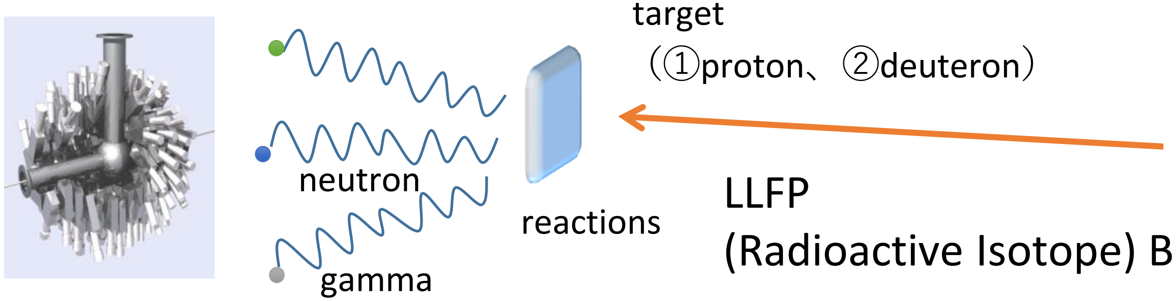
Total cross sections of spallation reaction could be expected larger than 1b.

Production cross section of each fragment gives half-life distributions of fragments.

RIBF provides a unique opportunity to get reaction data.

Nuclear Reaction Study via Inverse Reaction Method

Inverse reaction method to take nuclear reaction data



Stable/short-lived nuclei

target
（①proton、②deuteron）

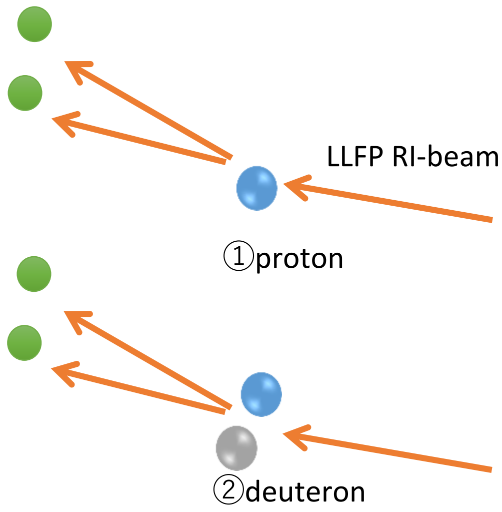
neutron

gamma

reactions

LLFP
(Radioactive Isotope) Beam

Spectrometer/detector
(PID of reaction products, etc)



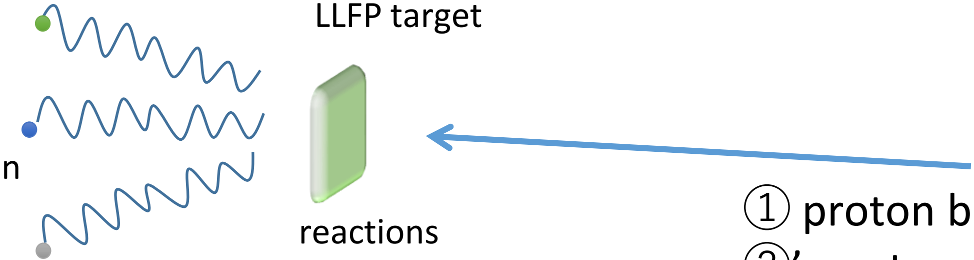
LLFP RI-beam

①proton

②deuteron
（neutron＋proton）

Nuclear transmutation setup

Stable/short-lived nuclei



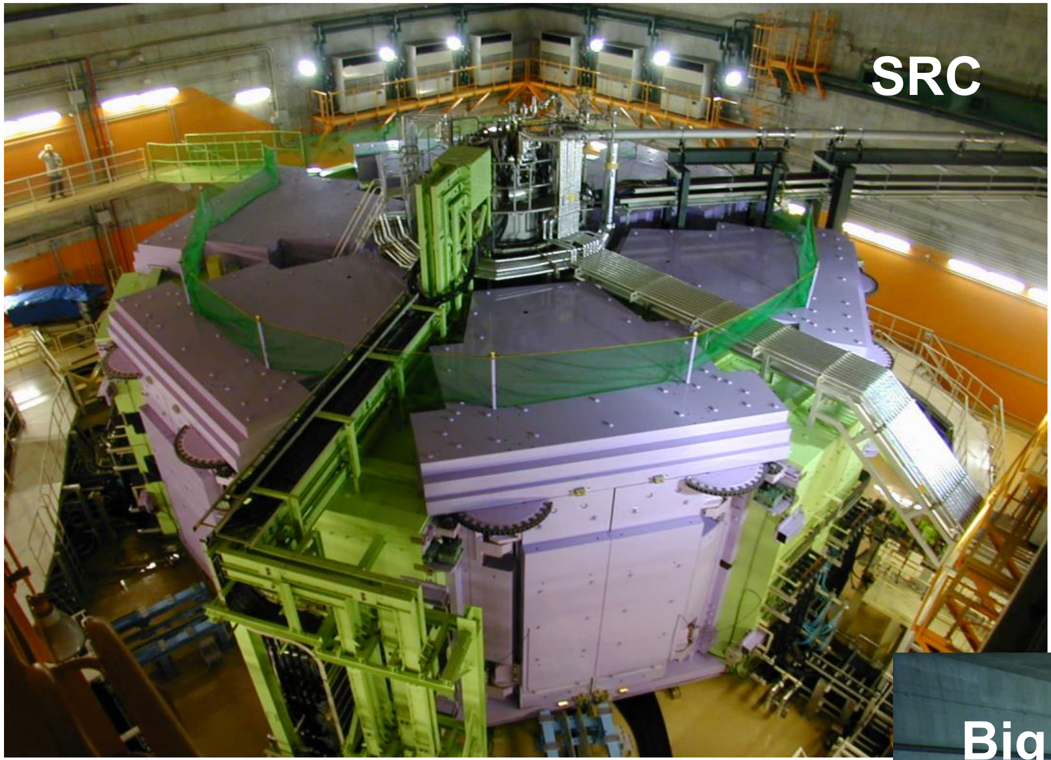
LLFP target

neutron

reactions

gamma

① proton beam
②’ neutron beam



World's First and Strongest
K2600MeV
Superconducting Ring Cyclotron

400 MeV/u Light-ion beam
345 MeV/u Uranium beam

World's Largest Acceptance
9 Tm
Superconducting RI beam Separator

~250-300 MeV/nucleon RIB

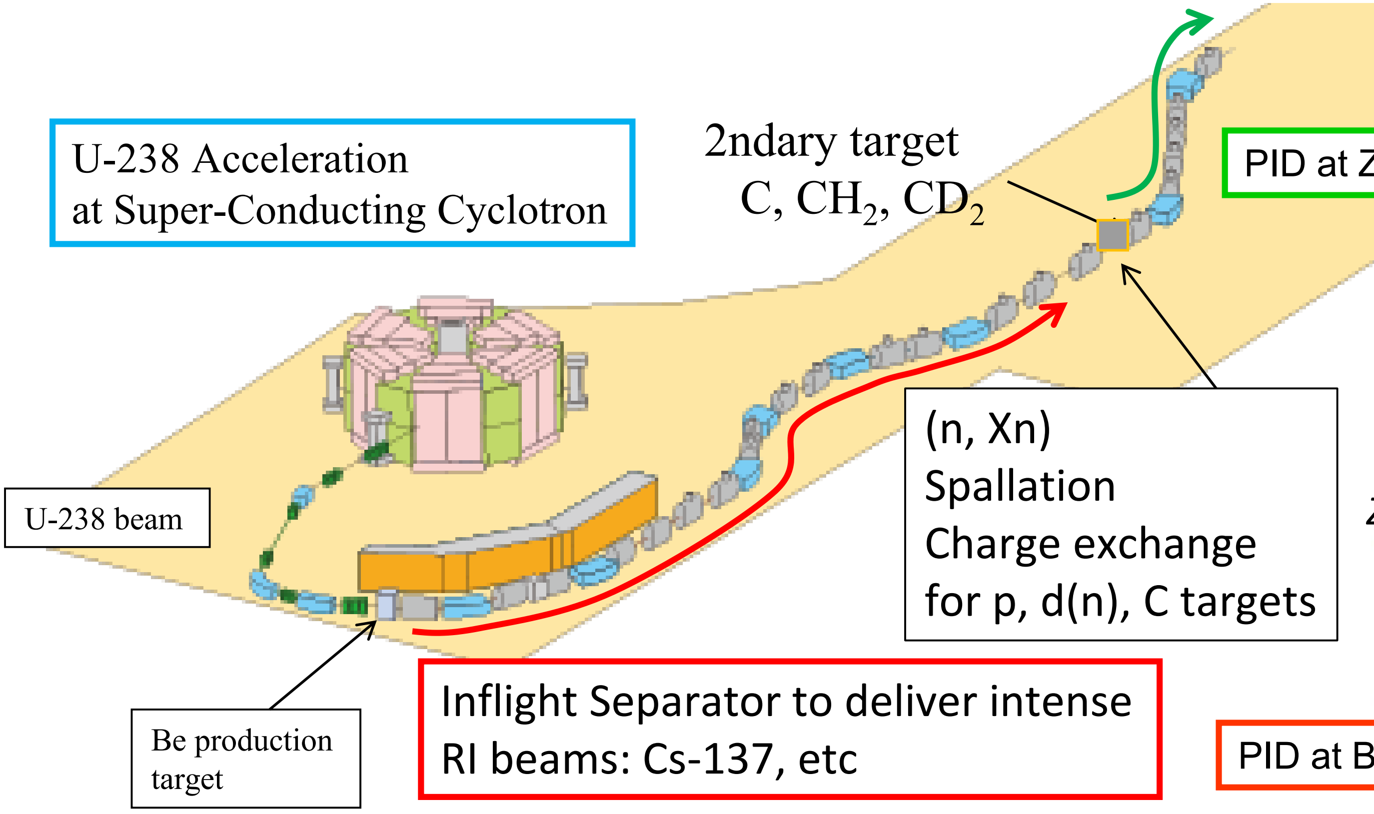
Transmutation for LLFP : The First Challenge

April, 2014

Beam species	Beam energy [MeV/u]	Intensity [/s/10pnA]	Purity [%]
^{137}Cs	186	1200	14
^{90}Sr	187	7100	28

ZeroDegree Spectrometer

PID for reaction products to determine reaction channels.



U-238 Acceleration
at Super-Conducting Cyclotron

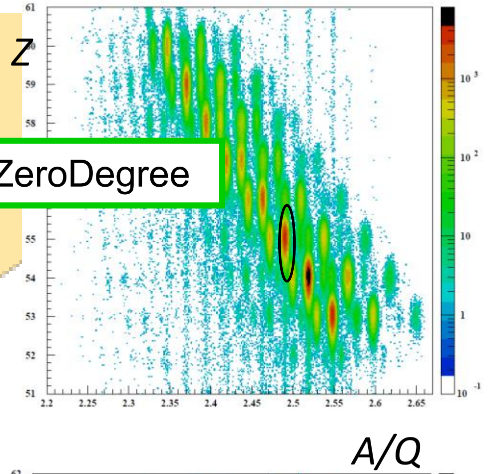
2ndary target
C, CH_2, CD_2

U-238 beam

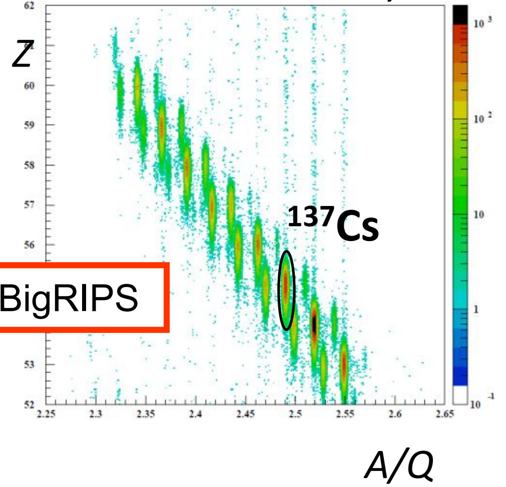
(n, Xn)
Spallation
Charge exchange
for p, d(n), C targets

Be production target

Inflight Separator to deliver intense RI beams: Cs-137, etc

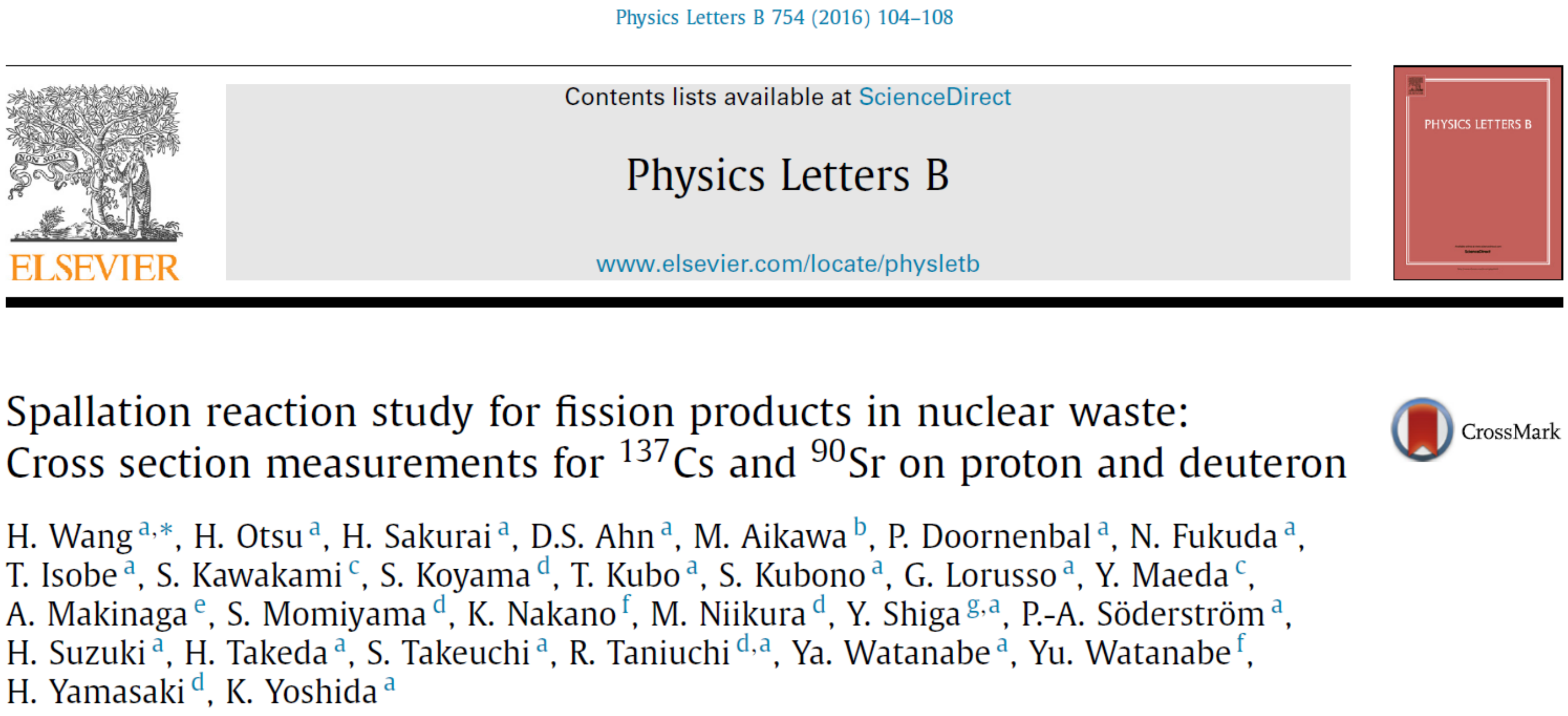


PID at ZeroDegree



PID at BigRIPS

RIKEN, UT, Miyazaki, Kyushu …



Physics Letters B 754 (2016) 104–108

Contents lists available at ScienceDirect

Physics Letters B

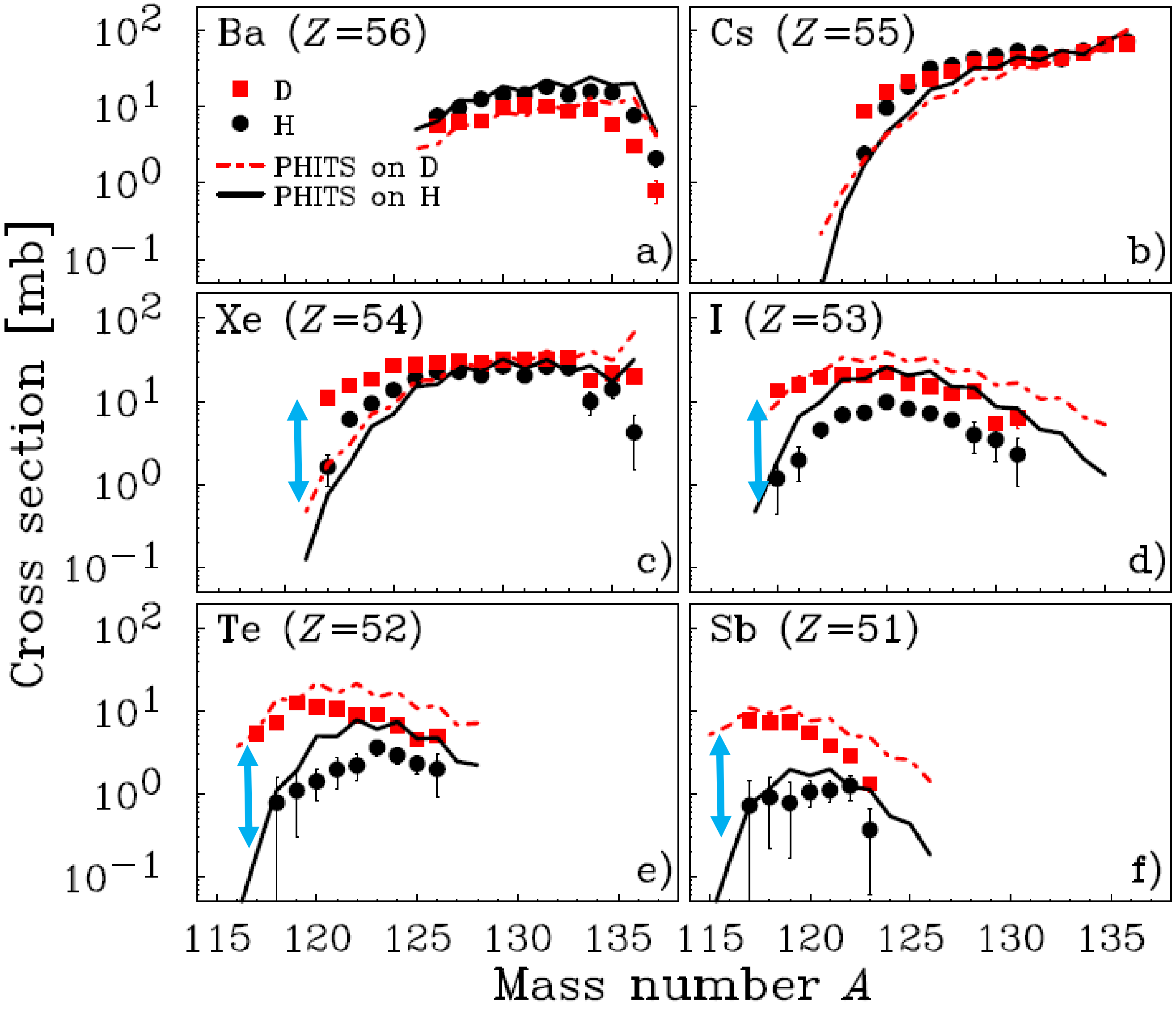
www.elsevier.com/locate/physletb

Spallation reaction study for fission products in nuclear waste: Cross section measurements for ^{137}Cs and ^{90}Sr on proton and deuteron

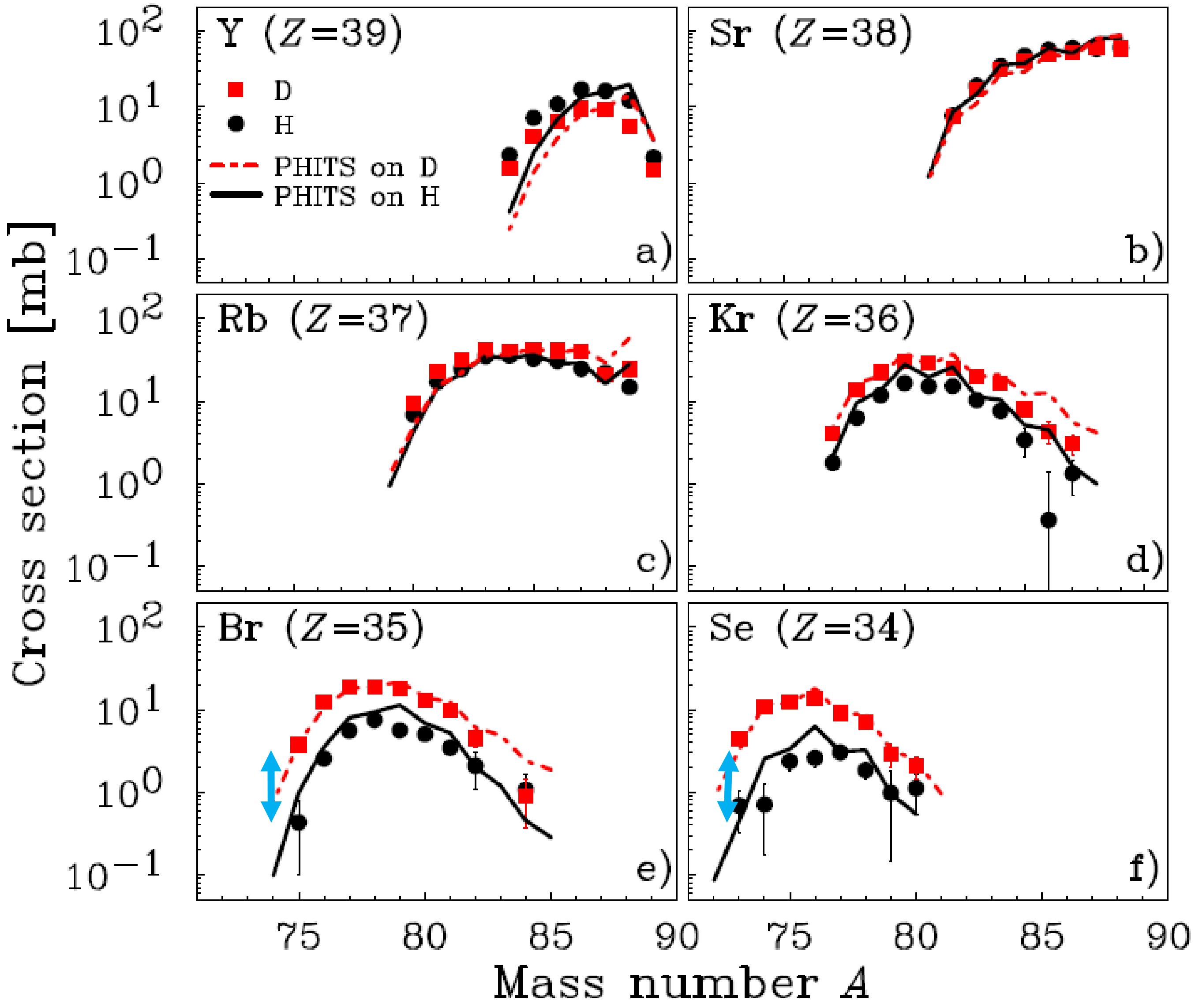
H. Wang[a,*], H. Otsu[a], H. Sakurai[a], D.S. Ahn[a], M. Aikawa[b], P. Doornenbal[a], N. Fukuda[a], T. Isobe[a], S. Kawakami[c], S. Koyama[d], T. Kubo[a], S. Kubono[a], G. Lorusso[a], Y. Maeda[c], A. Makinaga[e], S. Momiyama[d], K. Nakano[f], M. Niikura[d], Y. Shiga[g,a], P.-A. Söderström[a], H. Suzuki[a], H. Takeda[a], S. Takeuchi[a], R. Taniuchi[d,a], Ya. Watanabe[a], Yu. Watanabe[f], H. Yamasaki[d], K. Yoshida[a]

H. Wang et al, Physics Letters B 754, 104 (2016)

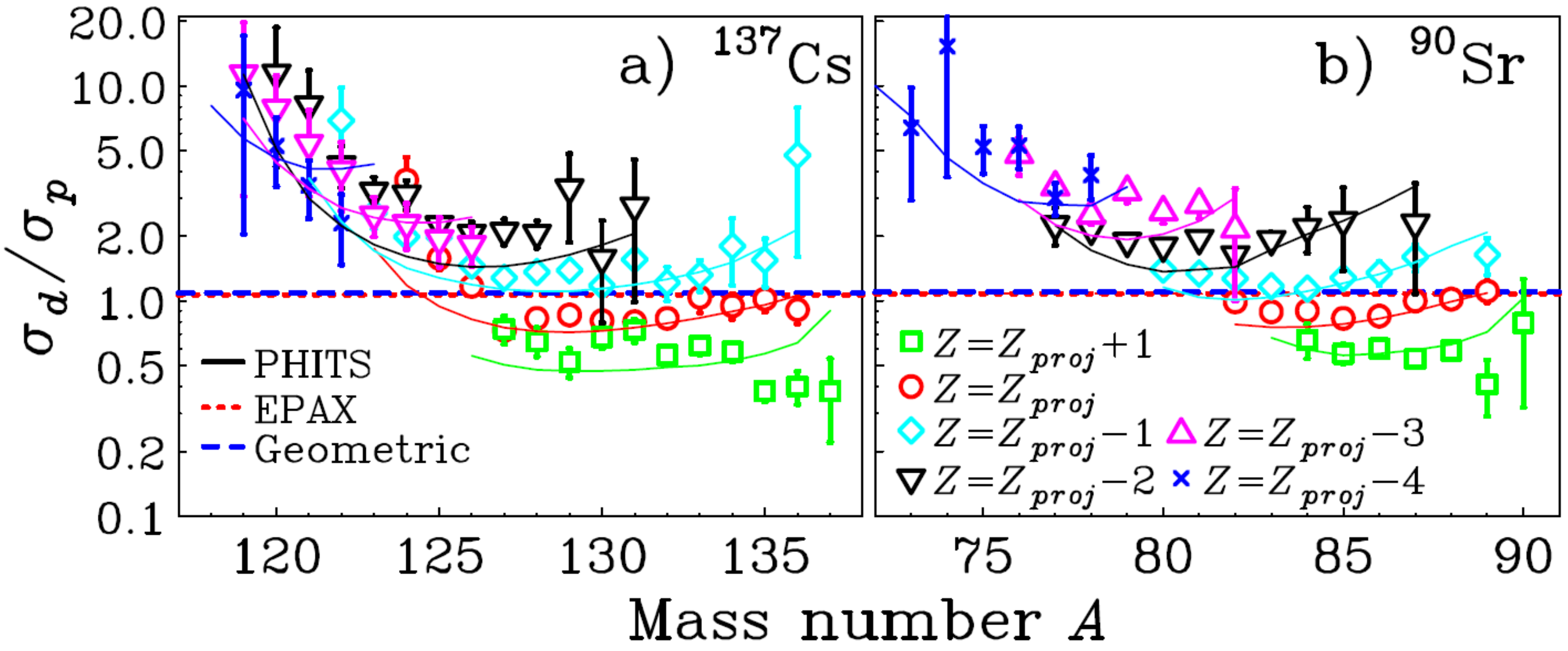
Cross section data ^{137}Cs + p, d



Cross section data ^{90}Sr + p, d



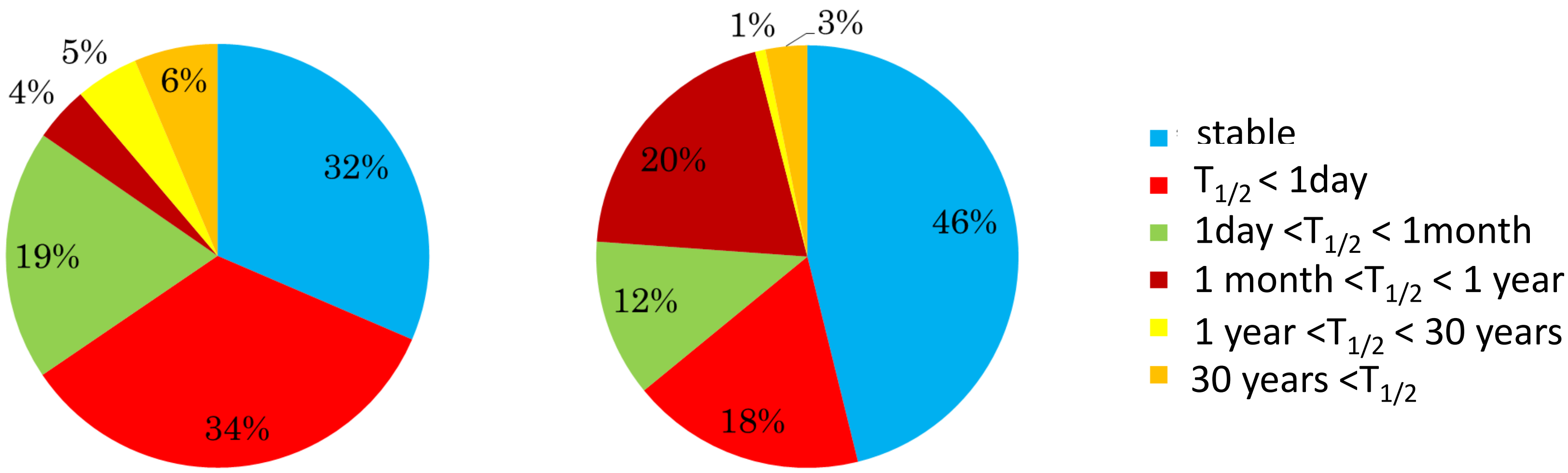
Comparison
between d-induced and p-induced reactions



Deuteron is a particle formed of proton and neutron, but not an in-coherent composite of proton and neutron !
It is hard to get "neutron"-induced components.

Deuteron gives higher energy deposits in targets

Half-life Distributions of Fragments



^{137}Cs + d

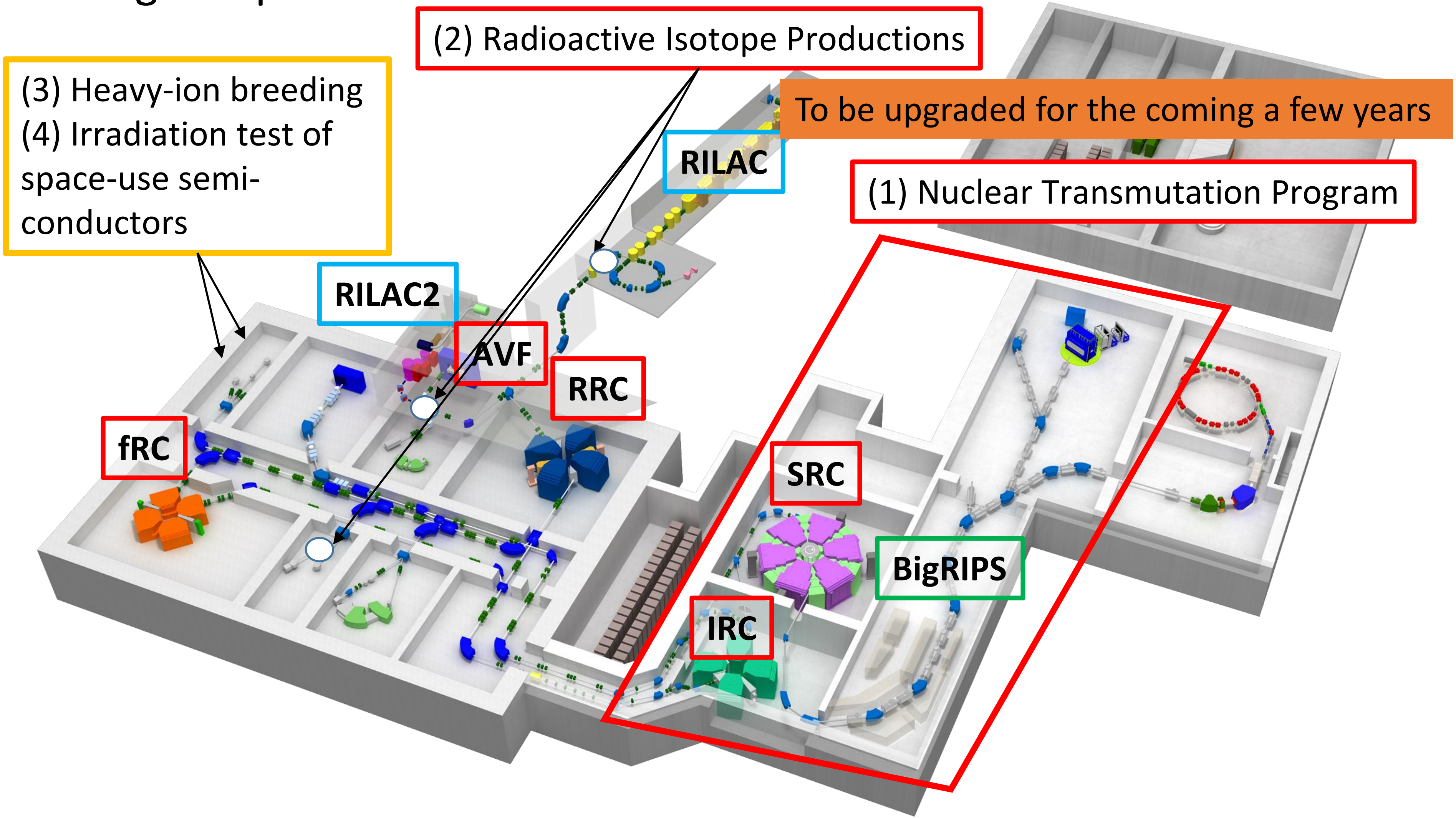
^{90}Sr + d

Fragments of which half-life is longer than 30 years are less than several percent.

Application Programs at RIBF

5 cyclotrons + 2 linacs

3 inflight separators



(2) Radioactive Isotope Productions

(3) Heavy-ion breeding

(4) Irradiation test of space-use semi-conductors

To be upgraded for the coming a few years

(1) Nuclear Transmutation Program

RIKEN RIs for application studies

Development of RI production technologies at AVF, RRC, and RILAC

- RI application studies in the fields of physics, chemistry, biology, medicine, pharmaceutical and environmental sciences
- Fee-based RI distribution to general public (FY2007–)
- Platform for short-lived RI distribution (FY2016–)

^{67}Cu for nuclear medicine

^{85}Sr: New product for fee-based RI distribution

^{211}At for nuclear medicine

^{262}Db, ^{265}Sg, and ^{266}Bh for SHE chemistry

Nuclides	Z	$T_{1/2}$	Accel.	Reactions	Research fields
^{7}Be	4	53.29 d	AVF	^{7}Li(p,n)	Ind.
^{28}Mg	12	20.91 h	AVF		Chem.
^{48}V					
^{48}Cr					
^{43,44m,44g,46,47,48}Sc	21	3.891 h, 58.6 h, 3.927 h, 83.79 d, 3.3492 d, 43.67 h	AVF	^{nat}Ti(d,X)	Phys., Med.
^{51}Cr	24	27.702 d	AVF	^{nat}Fe(d,X)	Phys.
^{52g}Mn	25				
^{54}Mn	25				
^{56}Mn					
^{55,56,57,58}					
^{59}Fe	26				
^{60}Co	27				
^{62}Zn	30				
^{64}Cu	29				
^{65}Zn	30	244.3 d	AVF	^{65}Cu(p,n) ^{65}Cu(d,2n)	Phys., Chem., Biol., Med., Pharm. sci., Ind., Environ. sci.
^{67}Cu	29				Pharm. sci., Med.
^{75}Se	34	119.779 d	AVF	^{75}As(p,n) ^{75}As(d,2n)	Biol., Med., Pharm. sci., Environ. sci.
^{85g}Sr				^{85}Rb(p,n) ^{85}Rb(d,2n)	Environ. sci. Chem.
^{88}Y	39	106.7 d	AVF	^{88}Sr(p,n) ^{88}Sr(d,2n)	Biol., Med., Pharm. sci. Chem.
^{85g}Zr*		7.86 min	AVF/RILAC	^{nat}Ge(^{18}O,xn)	Chem.
^{87g,88}Y	39	79.8 h, 106.65 d	AVF	^{nat}Zr(p,X) ^{nat}Zr(d,X)	Phys.
^{87m}Y	39	13.37 h	AVF	^{nat}Zr(d,X)	Phys.
^{88}Zr	40	83.4 d	AVF	^{89}Y(d,3n)	Chem.
^{89g}Zr	40	78.41 h	AVF	^{89}Y(p,n) ^{89}Y(d,2n)	Chem., Pharm. sci., Med.
^{89m}Zr*	40	4.18 min	AVF	^{89}Y(p,n) ^{89}Y(d,2n)	Chem.
^{88m,g}Nb*	41	7.8, 14.5 min	AVF/RILAC	^{nat}Ge(^{19}F,xn)	Chem.
^{90m}Nb*	41	18.81 s	AVF	^{90}Zr(p,n)	Phys.
^{90g,91m,92m,95m,95g,96}Nb	41	14.60 h, 60.86 d, 10.15 d, 86.6 h, 34.975 d, 23.35 h	AVF	^{nat}Zr(p,X) ^{nat}Zr(d,X)	Phys., Chem.
^{92m}Nb	41	10.15 d	AVF	^{92}Zr(p,n)	Chem.
^{90}Mo*	42	5.67 h	AVF/RILAC	^{nat}Ge(^{22}Ne,xn)	Chem.
^{93m}Mo*	42	6.85 h	AVF	^{93}Nb(p,n) ^{93}Nb(d,2n) ^{nat}Zr(α,X)	Chem.
^{93g,94g}Tc*	43	2.75 h, 293 min	AVF	^{nat}Nb(α,X)	Chem.
^{95}Zr	40	64.02 d	AVF	^{nat}Zr(p,X) ^{nat}Zr(d,X)	Phys.
^{97}Zr	40	16.91 h	AVF	^{nat}Zr(d,X)	Phys.
^{95m}Tc	43	61 d	AVF	^{95}Mo(p,n)	Environ. sci., Chem.
^{99}Mo	42	65.94 h	AVF	^{96}Zr(α,n)	Chem.
^{104m,g}Ag*	47	33.5 min, 69.2 min	AVF	^{nat}Pd(d,X)	Chem.

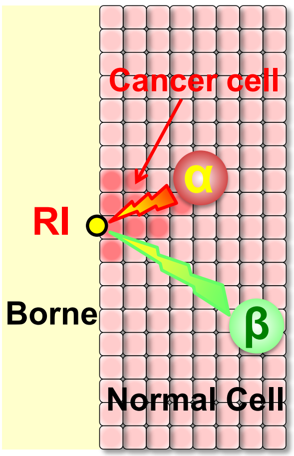
Nuclides	Z	$T_{1/2}$	Accel.	Reactions	Research fields
^{109m}Pd*	46	4.696 min	AVF	^{nat}Pd(d,X)	Chem.
^{141m}Nd*	60	62.0 s	AVF	^{141}Pr(d,2n)	Chem.
^{169,175,177}Yb	70	32.026 d, 4.185 d, 1.911 h	AVF	^{nat}Yb(d,X)	Phys.
^{177}Ta	73	56.56 h	AVF	^{nat}Hf(p,X)	Chem.
^{177}W*	74	135 min	AVF	^{nat}Hf(α,X)	Chem.
^{178a}Ta*	73	2.36 h	AVF	^{nat}Hf(d,X)	Chem.
^{179m}W*	74	6.40 min	AVF	^{nat}Ta(d,X)	Chem.
^{179m2,180m,181g}Hf	72	25.05 d, 5.5 h, 42.39 d	AVF	^{nat}Hf(d,X)	Phys.
^{180g}Ta	73	8.152 h	AVF	^{nat}Hf(d,X)	Phys.
Re*	75	2.44 min	AVF	^{nat}Ta(α,X)	Chem.
^{181}W	74	121.2 d	AVF	^{181}Ta(p,n)	Chem.
^{182}Ta	73	114.43 d	AVF	^{nat}Hf(α,X)	Chem.
^{185}Os	76	93.6 d	AVF	^{185}Re(p,n)	Chem.
^{188,189,191}Pt	78	10.2 d, 10.87 h, 2.802 d	AVF	^{nat}Os(α,X)	Med.
Pb	82	51.87 h	AVF	Tl(p,n)	Med.
Bi	83	6.243 d	AVF	^{206}Pb(p,n)	Pharm. sci., Med.
^{211}At					Med.
^{206}Fr*	87	15.9 s	RILAC	^{169}Tm(^{40}Ar,3n)	Phys., Chem.
^{209}Fr*	87	50.0 s	RILAC	^{197}Au(^{18}O,6n)	Phys., Chem.
^{214}Ac*	89	8.2 s	RILAC	^{197}Au(^{22}Ne,5n)	Phys., Chem.
^{245}Fm*	100	4.2 s	RILAC	^{208}Pb(^{40}Ar,3n)	Phys., Chem.
^{255}No*	102	3.1 min	AVF/RILAC AVF	^{238}U(^{22}Ne,5n) ^{248}Cm(^{12}C,5n)	Phys., Chem.
^{255}Lr*	103	22 s	RILAC	^{209}Bi(^{48}Ca,2n)	Phys.
^{257}Lr*	103	0.646 s	AVF	^{248}Cm(^{14}N,5n)	Phys.
^{259}Lr*	103	6.3 s	AVF	^{248}Cm(^{15}N,4n)	Phys.
^{261a,b}Rf*	104	68, 1.9 s	AVF/RILAC	^{248}Cm(^{18}O,5n)	Phys., Chem.
^{262}Db*					Chem.
^{265a,b}Sg*	106	8.5, 14.4 s	RILAC	^{248}Cm(^{22}Ne,5n)	Phys., Chem.
^{266}Bh*		s	RILAC	^{248}Cm(^{23}Na,5n)	Phys., Chem.
Multitracer	<22		RRC	^{nat}Ti(^{14}N,xnyp)	Chem., Biol., Med., Pharm. sci., Ind., Environ. sci.
Multitracer*	<29		RRC	^{nat}Cu(^{14}N,xnyp)	
Multitracer	<47		RRC	^{nat}Ag(^{14}N,xnyp)	
Multitracer	<72		RRC	^{nat}Hf(^{14}N,xnyp)	
Multitracer*	<73		RRC	^{nat}Ta(^{14}N,xnyp)	
Multitracer	<79		RRC	^{197}Au(^{14}N,xnyp)	
Multitracer*	<83		RRC	^{209}Bi(^{14}N,xnyp)	

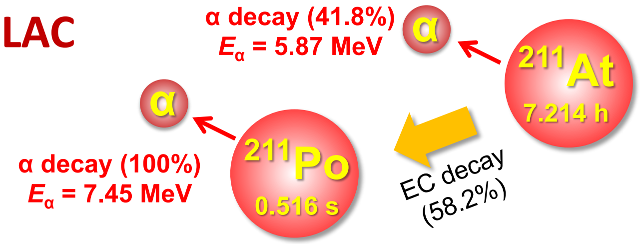
* RIs produced with the gas-jet system.

Production of promising therapeutic ^{211}At using ^{209}Bi(α,2n)^{211}At at AVF

Toward mass production of ^{211}At with 0.5 mA α beam from RILAC

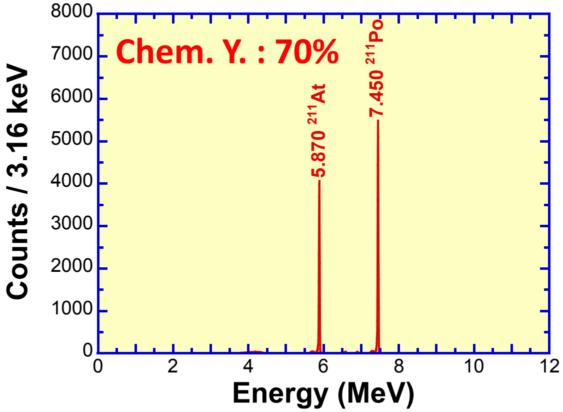
- Short range of α particle
- High LET
- Large cytotoxic effect, small effect for normal cell
- Effective for disseminative, blood, and spread cancers, and small cancer left after operation



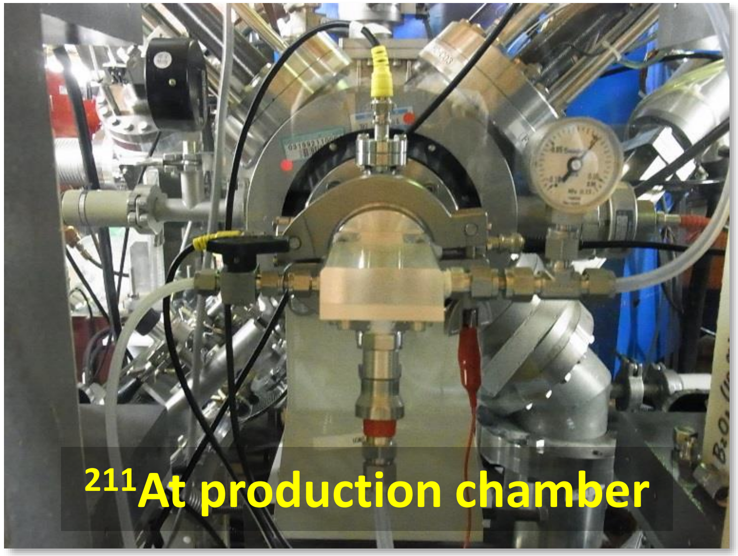


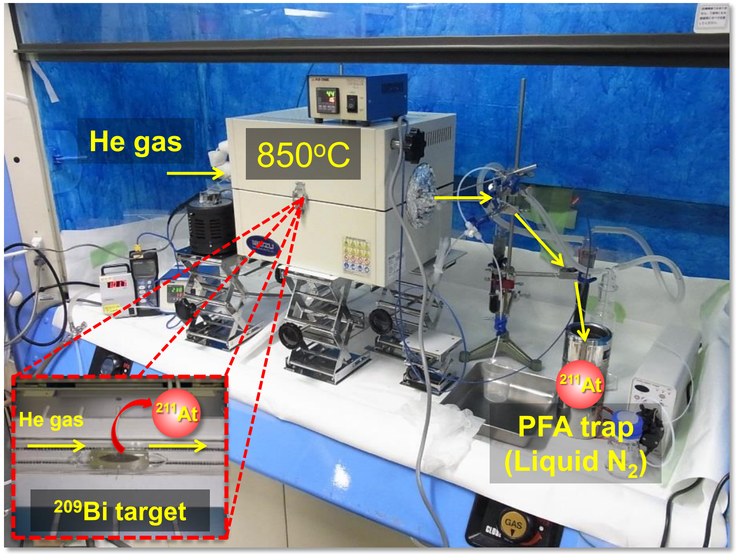
→ R&D of novel ^{211}At medicines in collaboration with Biofunctional Synthetic Chemistry Laboratory, Synthetic Cellular Chemistry Laboratory, RIKEN Center for Life Science Technologies, and RIKEN Innovation Center

α-ray spectrum of ^{211}At



H. Haba, J. Part. Accel. Soc. Jpn. **12**, 206 (2015).





Summary

RIBF has started a new project to obtain LLFP nuclear reaction data via inverse reaction method.
A bunch of reaction data are being obtained.
A conceptual design for accelerator transmutation systems is being discussed under collaboration of domestic universities/institutes.
e.g. Cyclotron, Linac, FFAG…

A project has started to deliver At-211 to chemists and biologists for development of new nuclear medicine.