液体金属強力中性子源と中性子科学利用の展望

H. Horiike

Guest Professor, Osaka University

T.Okita and E. Hoashi

Osaka University, Grad. School of Engineering, Suita City, Osaka

H.Kondo

National Institutes for Quantum and Radiological Science and Technology (Mitsubishi Heavy Industries Ltd.)

T. Kanemura FRIB, Michigan State University

目次

- 液体金属リチウム高速流の研究開発について
- 大阪大学と大洗のリチウム装置での開発の概要
- Liベース中性子源の科学応用の可能性
  1. FRIBでの電子ストリッパー
  2. A-FNS
  3. 超長半減期核種の核変換処理の可能性
  4. 医療応用：BNCT
- まとめ

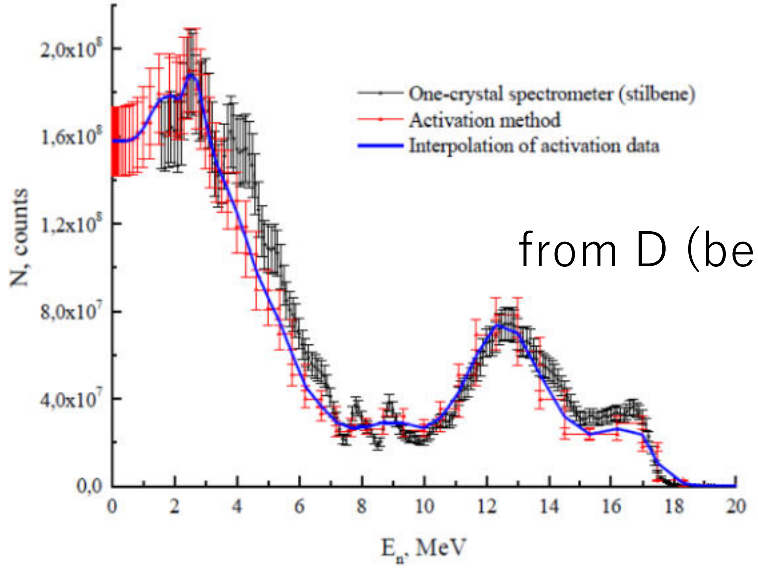
INTRODUCTION

- Liquid lithium is a candidate material for a target of intense **neutron source**, **heat transfer medium** in space engines and **charge stripper** for FRIB. For IFMIF development, liquid lithium free surface test facilities were constructed at Osaka Univ. and JAEA, and promising results have obtained as design data bases for the IFMIF and A-FNS. The latter is planned to construct in the mid 20's in Rokkasho Aomori as one of BA project, and will be used for high energy high dpa neutron tests for DEMO components.
- A good application of IFMF technology is in **transmutation** processing of long life fission products. These elements contain isotopes of several atomic numbers and only one isotope among them has very long half decay time. These could be transmuted to stable or short decay elements through high energy neutron irradiation. IFMIF could be used to measure the cross sections relevant to these reactions so as to precisely design the system.
- In medical application of **BNCT**, thermal neutrons with least energetic neutrons and gamma rays are required, so as to avoid unnecessary doses to a patient. This is enabled by lithium target irradiated by protons at 2.5MeV, since $^{7}Li(p, n)^{7}Be$ is a threshold reaction at 1.88MeV. Here protons penetrate into Li by 0.25mm with dissipating beam energy there. Thus a thin film flow at high velocity is important for stable operation of the treatment. Neutrons generated at 0.6MeV are gently moderated to epithermal range while suppressing accompanying gamma ray and energetic neutrons minimum by a moderator assembly.
- For these application, high current proton beam is necessary. A candidate is a tandem accelerator, which is 1.25 times higher energy than the ITER neutral beam. This system will enable extraction of an ampere class beams at several MeV with H or D with using market power supplies.

$^{7}Li(d,n)^{8}Be$ and $^{7}Li(p,n)^{7}Be$

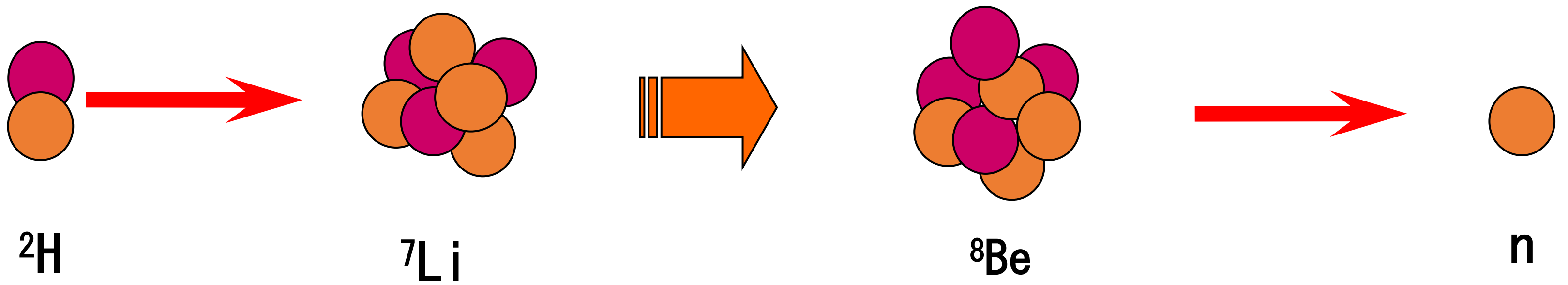
$^{7}Li(d,n)^{8}Be$ by 2.9MeV D beam

from $^{8}Be^{*}$(excited)

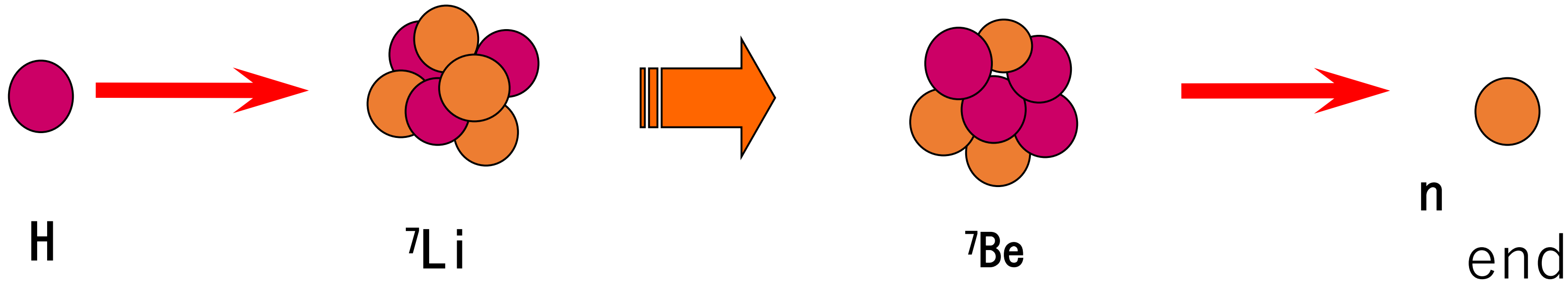


from D (beam)

exoergic reaction



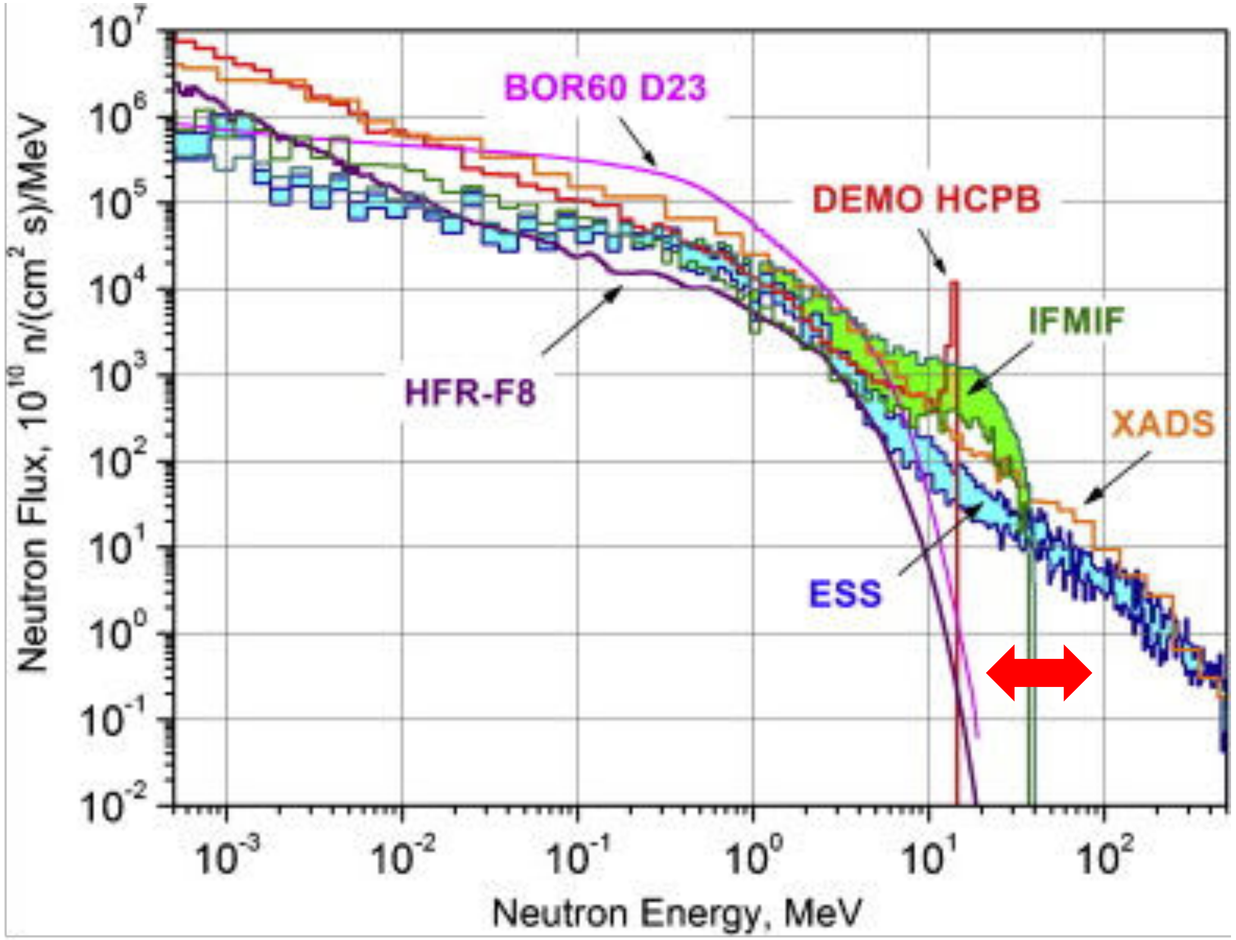
High energy neutron source



Thermal neutron source

endoergic reaction

Neutron distribution of IFMIF and a fission reactor



Neutron fluxes are higher by 2 to 3 orders at 10MeV than those in fission reactors.

The upper slope of the distribution(green curve) is able to vary with the beam energy.

Profile selectable by beam

I. Lithium system R&Ds for IFMIF

1) A small experimental Li test loop at Osaka University mid 80's in same period with FMIT test

2) A small natural convective Li heat transfer test equipment under strong magnetic field in Osaka 1993

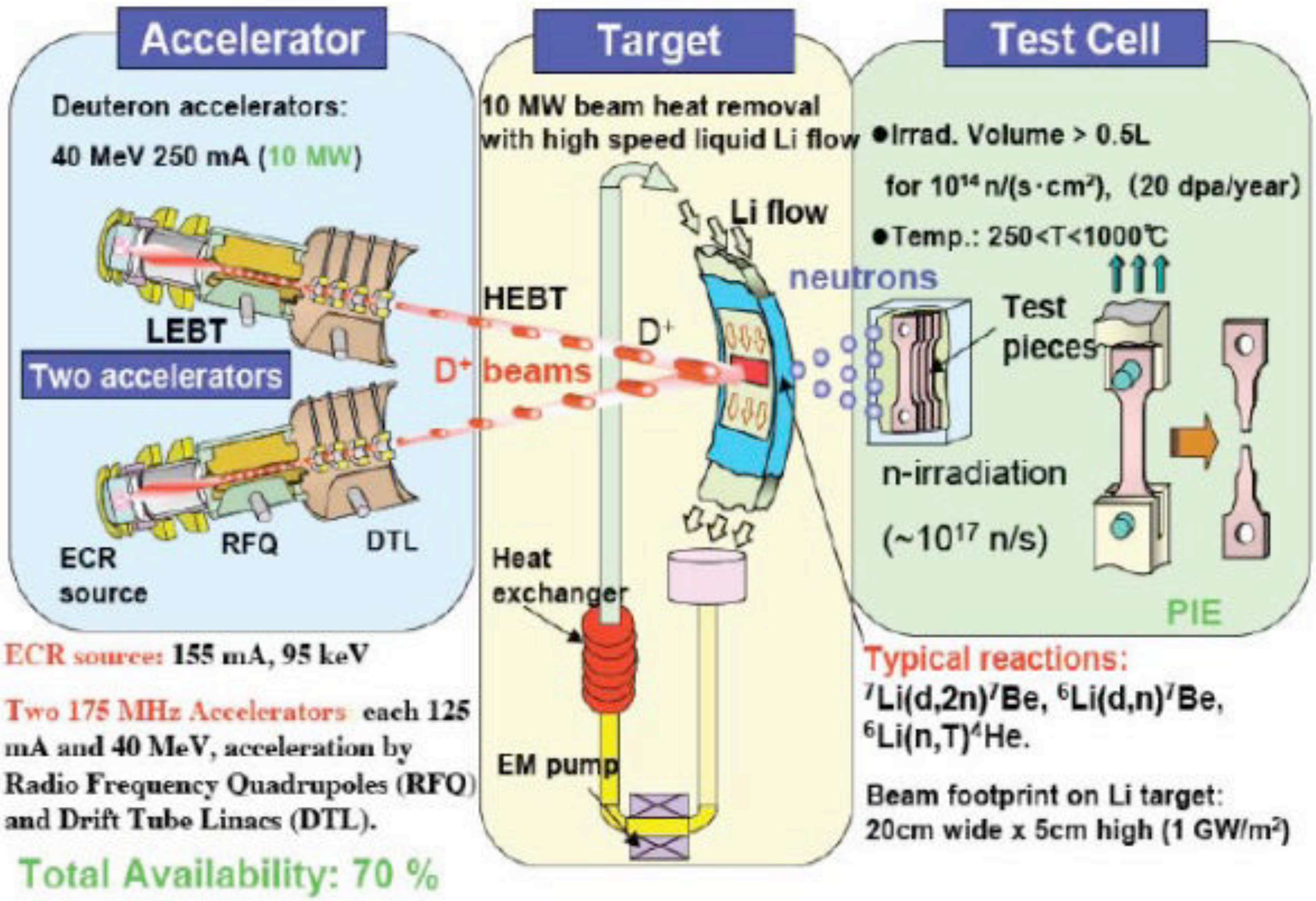
3) Forced convective heat transfer test loop under magnetic field 1997

4) High speed free surface experiment under KEP project 2000

5) ELEL facility constructed at JAEA and experiment under IFMIF-EVEDA 2011

6) Osaka Univ. loop modification for A-FNS design.

IFMIF Schematics

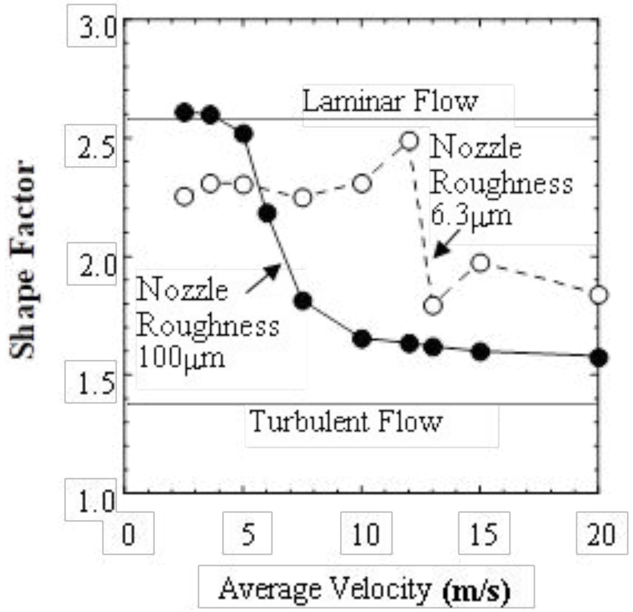


- D beams of 100-300mA at 30-40MeV
- Lithium flow of **25mm thick** at 15-20m/sec in 10^{-3}Pa
- Temp. controlled test cell units
- Handling high activated Li

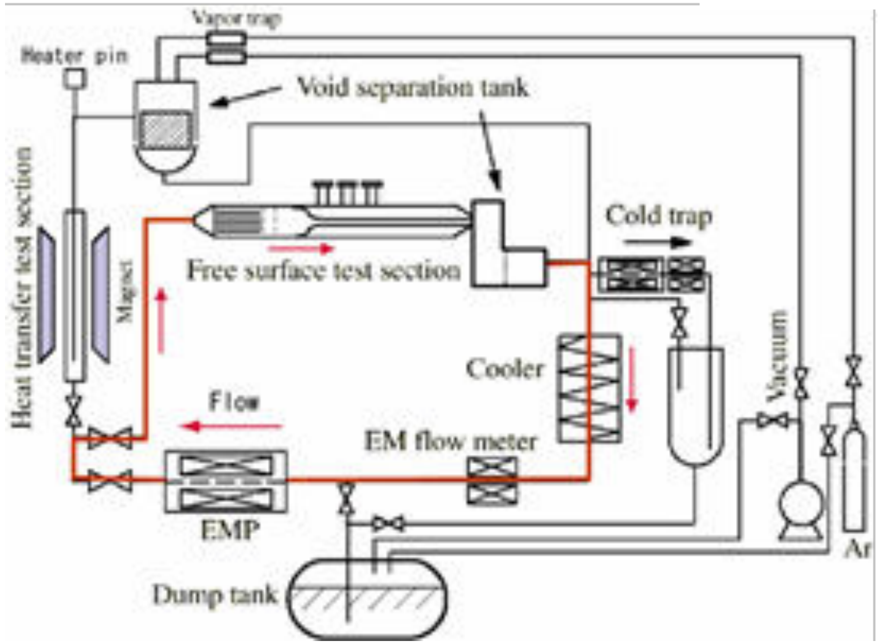
Application
- material test
- blanket study
- neutron science
- transmutation
- cancer treatment

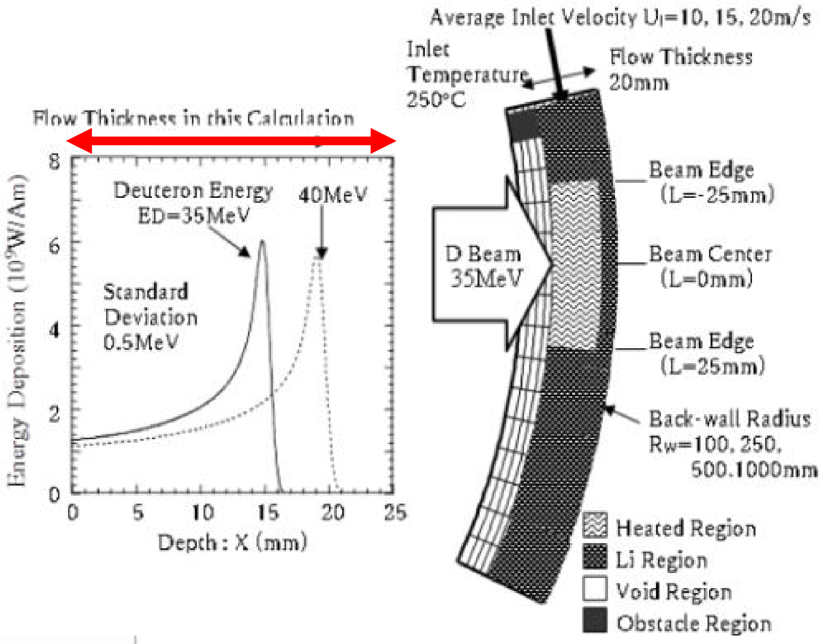
Li Fluid Dynamic Study at Osaka

Water simulation experiments were conducted at JAERI and showed importance of surface roughness inside of the nozzle.

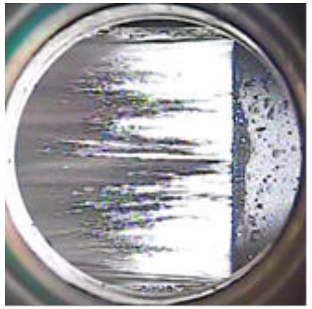


Li experimental facility was constructed to Osaka Univ loop



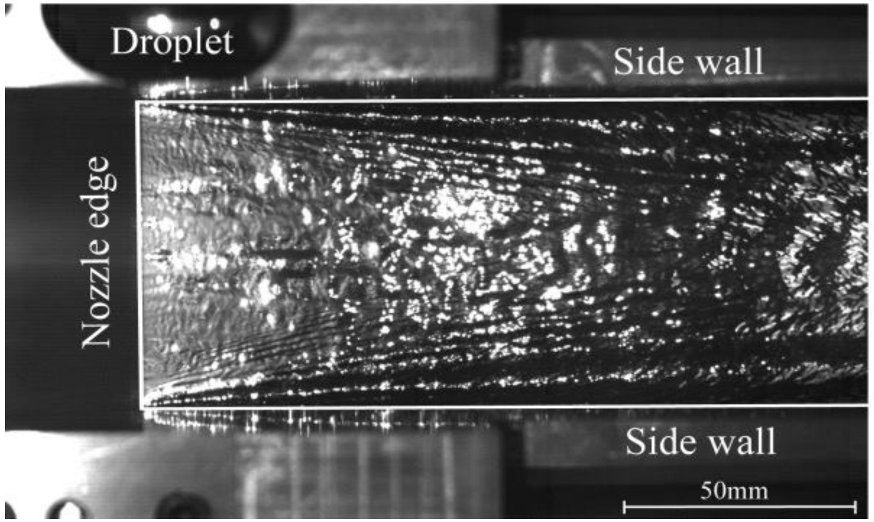


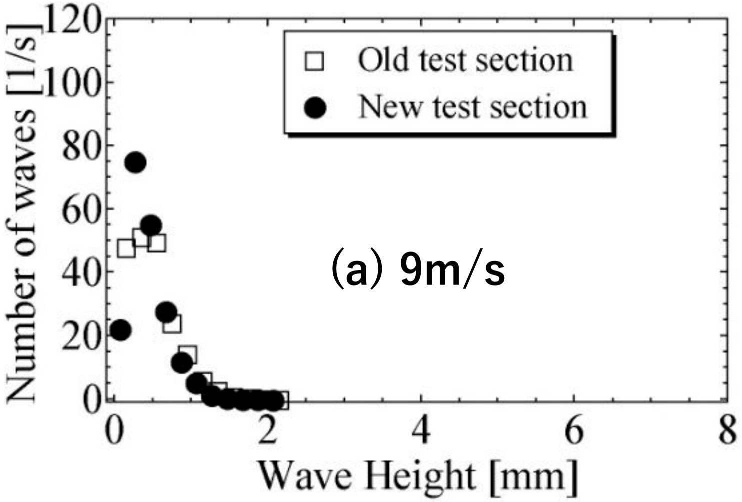
Lithium free surface flow of 15m/sec 2003

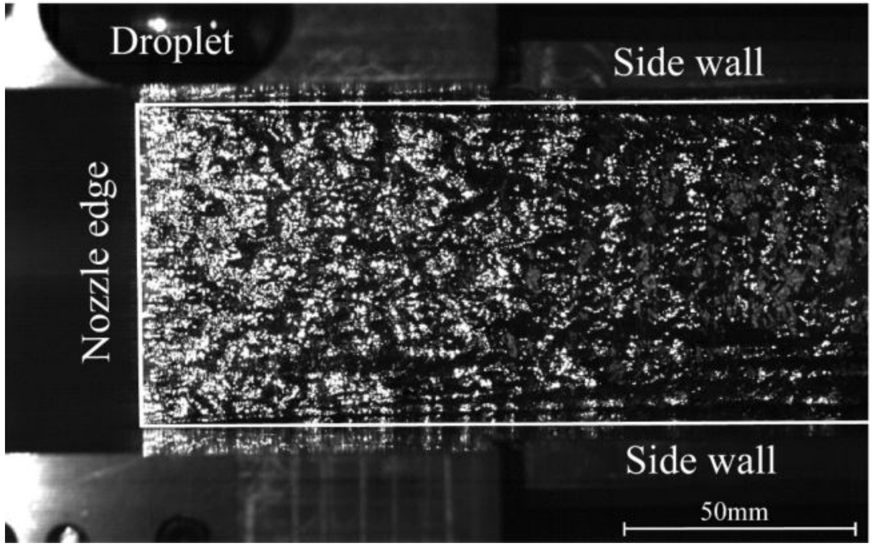


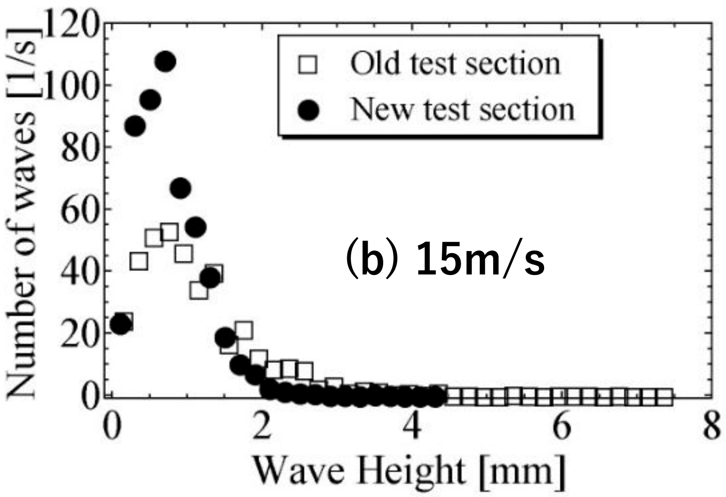
Li experimental facility at Osaka Univ.

Surface wave measurements 2





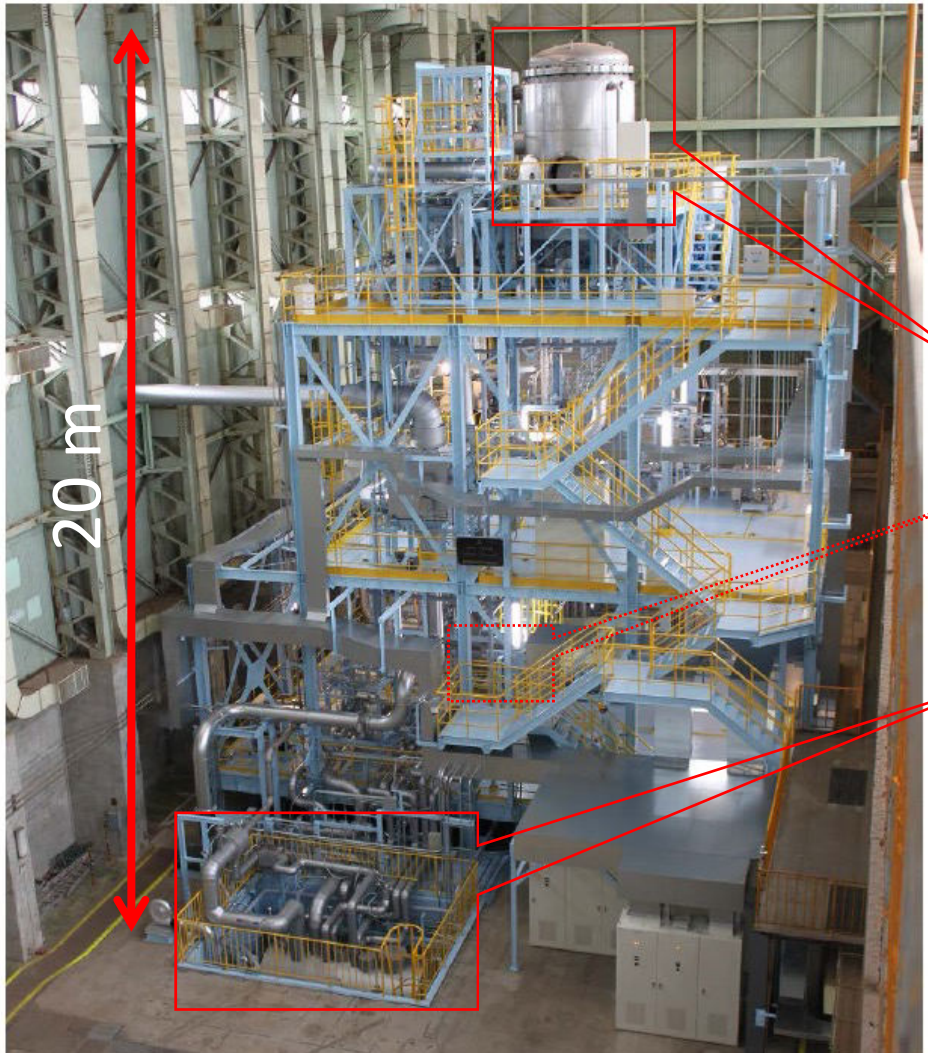




- With changing the nozzle, effect of nozzle sharpness was tested and found to be almost nothing.

ノズル　10mm厚 70mm幅

IFMIF/EVEDA Lithium Test Loop



Scale: 20m × 20m × 20 m
(Three stories and underground pit

< Components>

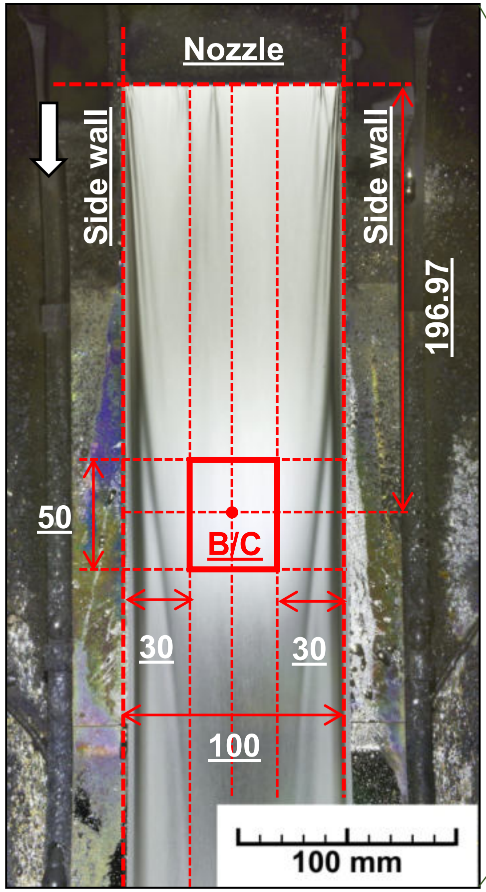
(1) Target Assembly

(2) EMP･EFM
Flow rate: 3000 l/min

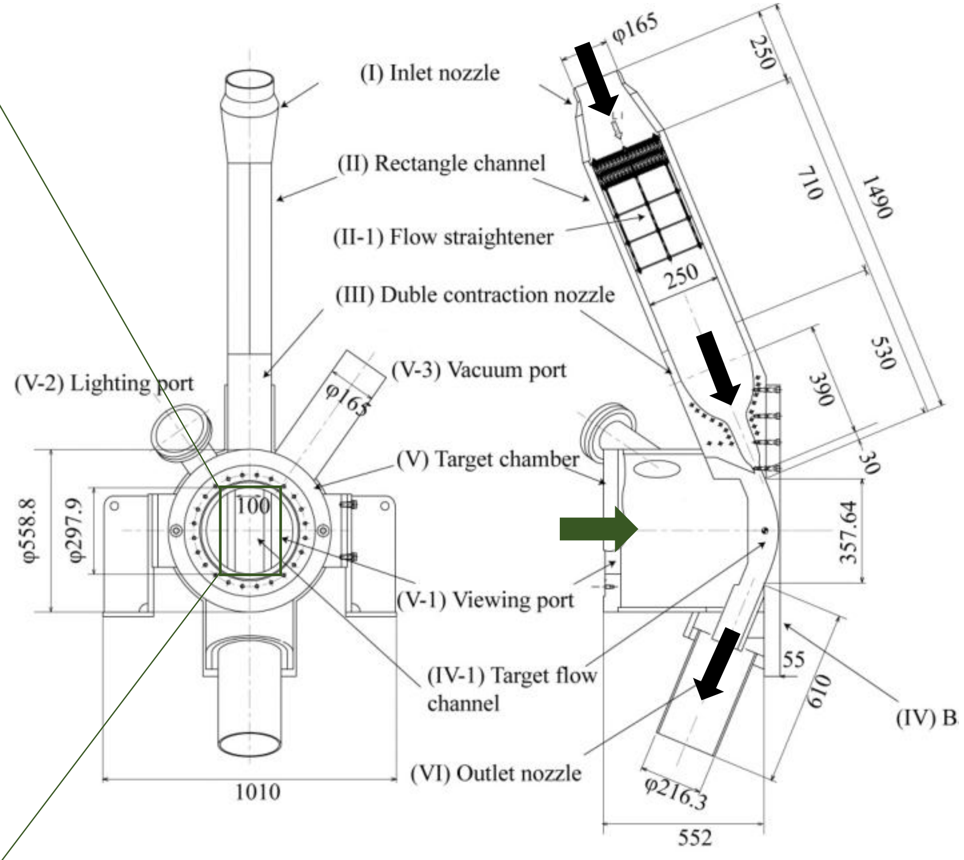
(3) Dump tank
Inventory: 2.5 ton (5000 l)

Largest Facility ever completed

Li target satisfies the Rated values



V = 15 m/s, P = 1 x 10^{-3} Pa
T = 250 °C



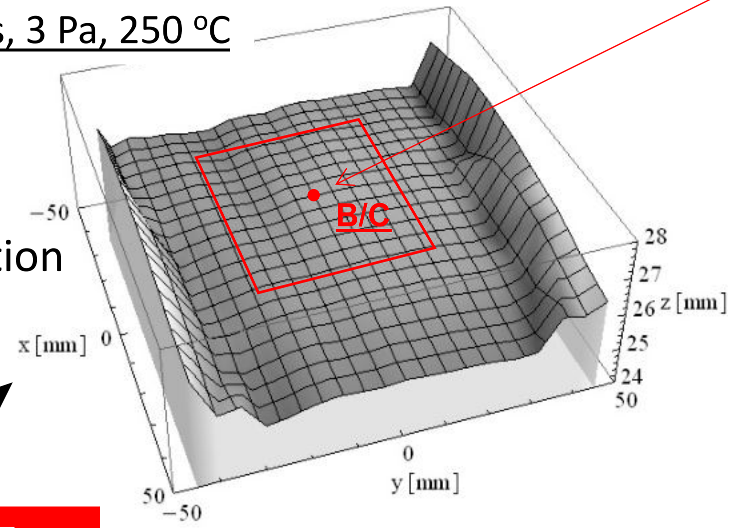
＊Width are reduced to 1/2.6

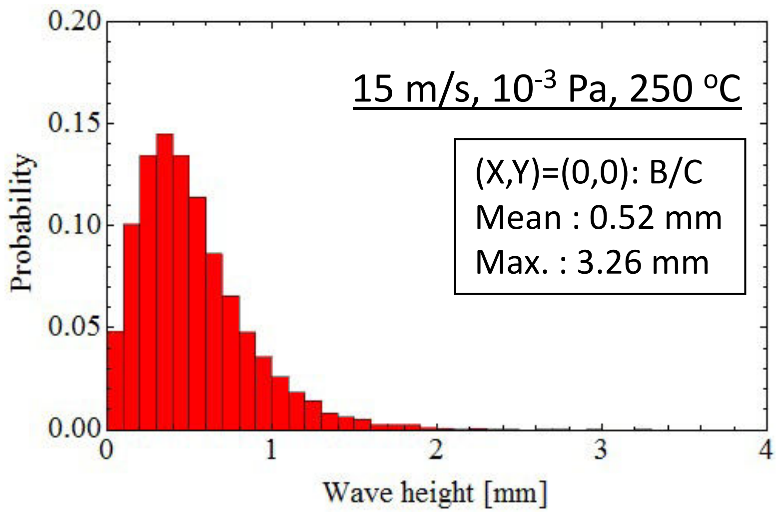
ノズル:
25mm厚
100mm幅

Li Flow is very stable and smooth

Measuement with using a Laser Probe

3D shapes are obtained（time average)	Surface waves are the same with previous ones





結果

Li flow depth:
- Uniformity（P-P）: **0.16 mm**

Wave amplitude (a half of height) of **99.7 % is** less than **1 mm**.
The same conclusion was confirmed again

Li target is stable
We can go to A-FNS & IFMIF

Summary for Lithium Flow Dynamics

- Lithium high speed flow in open channel has been studied and developed by IFMIF-KEP at Osaka University followed by IFMIF-EVEDA at JAEA Oarai.
- Systematic works over 20yr has revealed its potency in high heat flux handling under high vacuum environment.
- Precise measurement on the surface shapes has shown its **flatness** with small waves of random nature, and thus constructed facility was found to be very **suitable as the high energy beam target.**
- Basic fluid-dynamical properties of Li were found to obey those for existing fluid dynamics established with water and other fluids.

II. 電子ストリッパーへのLi流の応用

FRIB

Development of a Liquid Lithium Charge Stripper for FRIB

F. Marti[a], P. Guetschow[a],
Y. Momozaki[b], J. A. Nolen[a,b], C. B. Reed[b]
[a] MSU, [b] ANL

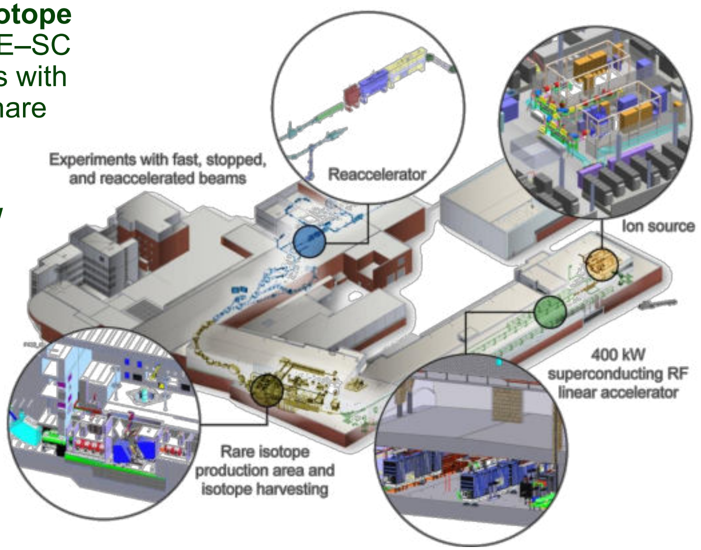
MICHIGAN STATE UNIVERSITY U.S. DEPARTMENT OF ENERGY Office of Science

This material is based upon work supported by the U.S. Department of Energy Office of Science under Cooperative Agreement DE-SC0000661, the State of Michigan and Michigan State University. Michigan State University designs and establishes FRIB as a DOE Office of Science National User Facility in support of the mission of the Office of Nuclear Physics.

FRIB(USA), RIBF(理研)は重元素イオンの高速度衝突実験により新元素(Rare Isotopes)を見つけるプロジェクトで、究極の重イオンビームは高速のウランビーム

What is FRIB?

- The **Facility for Rare Isotope Beams** is funded by DOE–SC Office of Nuclear Physics with contributions and cost share from Michigan State University
- Key features are 400 kW beam power for all ions and E/A> 200 MeV/u
- Separation of isotopes in-flight provides
  - Fast development time for any isotope
  - All elements and short half-lives
  - Fast, stopped, and reaccelerated beams



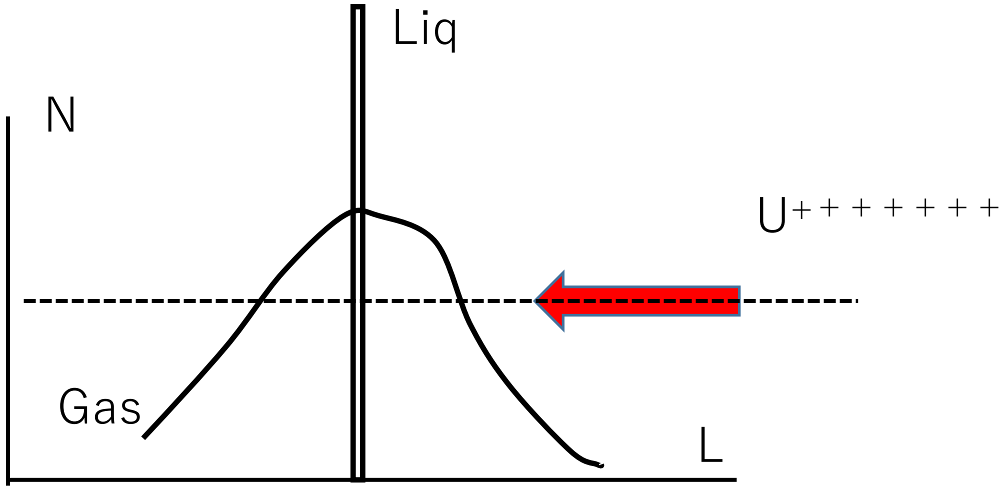
FRIB Facility for Rare Isotope Beams
U.S. Department of Energy Office of Science
Michigan State University

F. Marti, September 2015 HIAT 2015, Slide 3

ウランを効率良く加速する上で電荷数の増加は非常に重要．電子剥離と再結合抑制が重要

Electron Stripperの機能

- He ガスを使う方法と液体金属リチウム流による方法がある
- C, Be板はビーム熱負荷が大きくて使用できない



Heガスでの剥離：理研

$<U^{+62}$

排気が困難

液体Li：MSU

$<U^{+70\sim+75}$

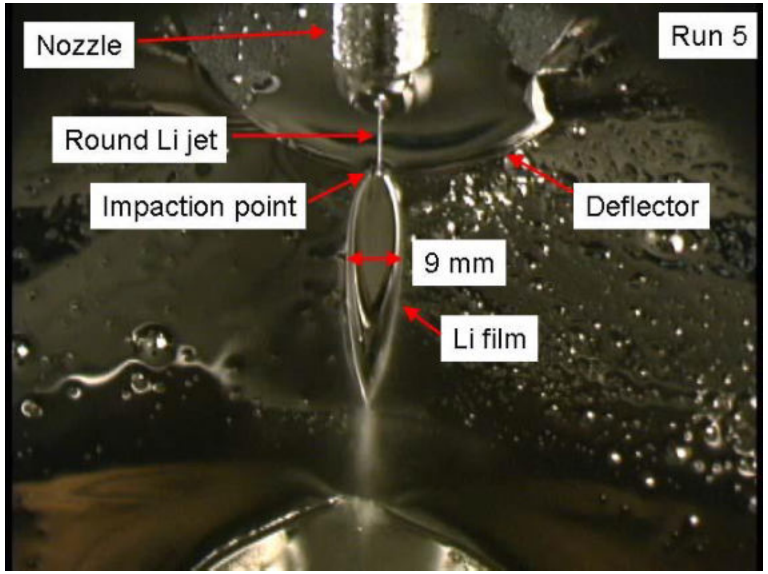
Li取扱いが大変

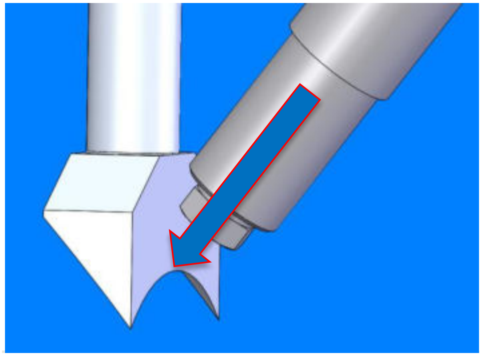
厚さ$<20\mu m$, 流速$<60 m/sec$

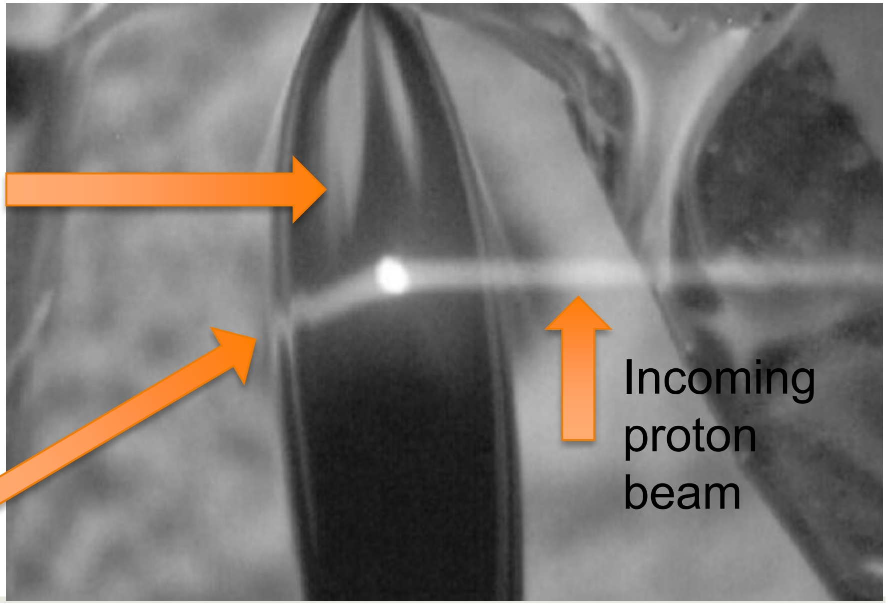
Gasの場合は剥離したイオンが下流で電子と再結合する

液体 固体の場合は再結合が小さい

Li薄膜流の生成



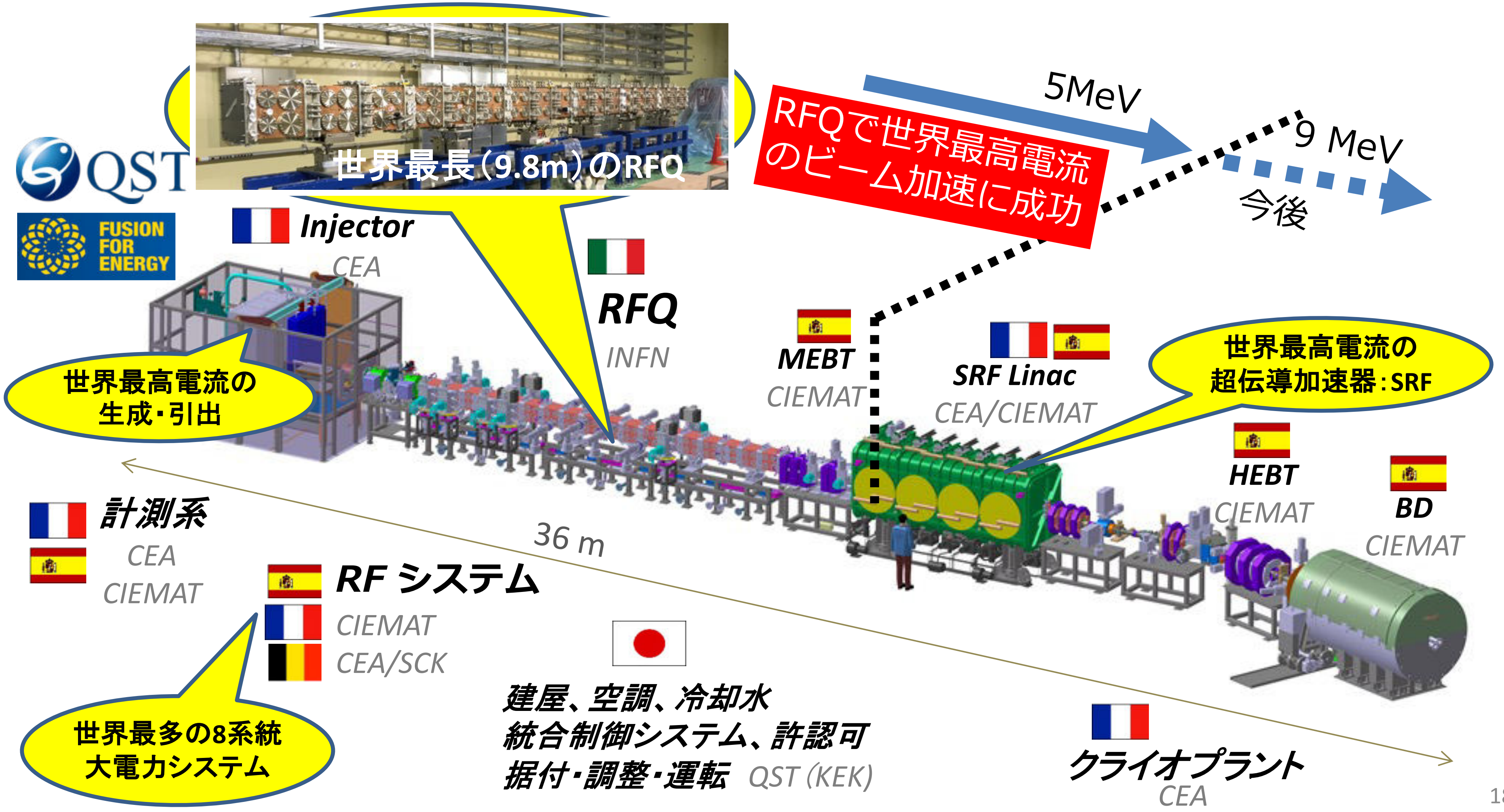




細いノズルから噴出するリチウム流を平板に当てて、反射して広がる薄膜を利用
膜厚が薄く、流速が上げられる. Li装置は試運転段階
今後のウランビームでの実験に期待

III. IFMIF原型加速器の開発

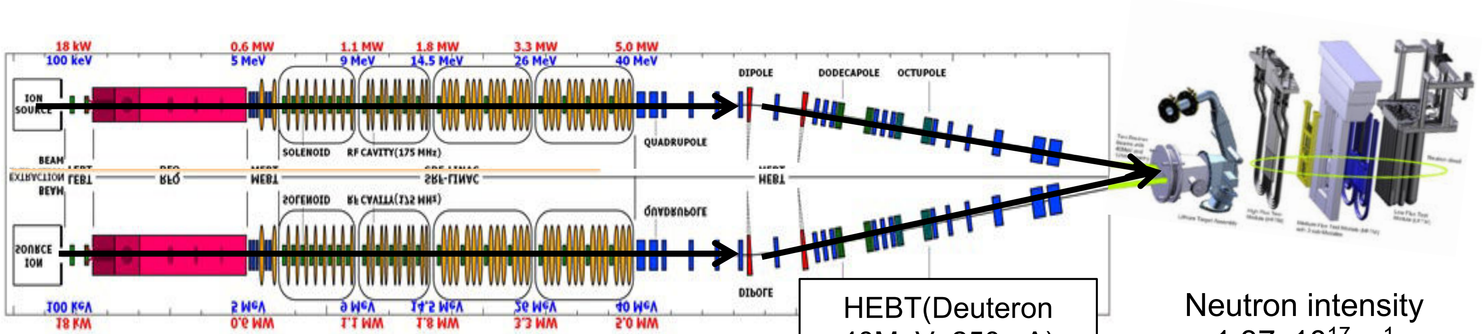
・目標：**重陽子ビーム、125mA、9MeV、連続加速の技術的な見込みを得る**
・実施体制：日本（QST)、欧州（F4E）のもと、**各国研究機関が機器調達し六ヶ所研で組立・試験**。（日本ではKEK「高エネルギー加速器研究機構」も参画）



IFMIF and A-FNS

IFMIF irradiates fusion materials

Basing on the results obtained so far, these facilities turned to reality



Injector RFQ SRF

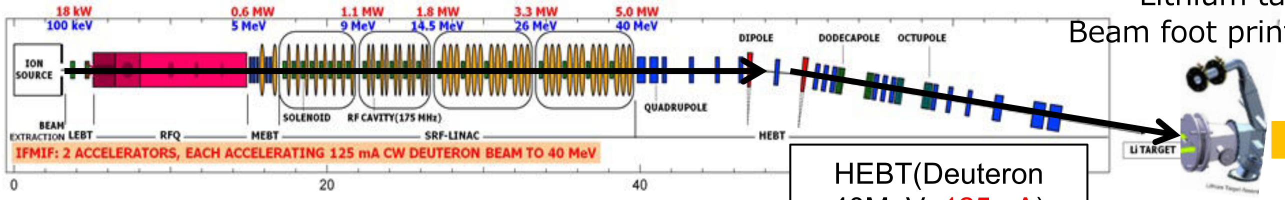
HEBT(Deuteron 40MeV, 250mA)

Neutron intensity $1.37 \times 10^{17}\ s^{-1}$

A-FNS

Single accelerator 125mA at present

Lithium target
Beam foot print 100cm²



HEBT(Deuteron 40MeV, 125mA)

Irradiation test on Fusion materials and general applications

Neutron intensity $6.8 \times 10^{16}\ s^{-1}$

Industrial and medical application possible

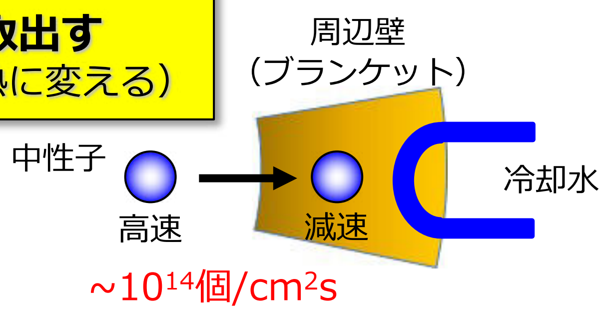
Flexible for irradiation module design

核融合中性子源：核融合炉材料の耐久性評価にむけて

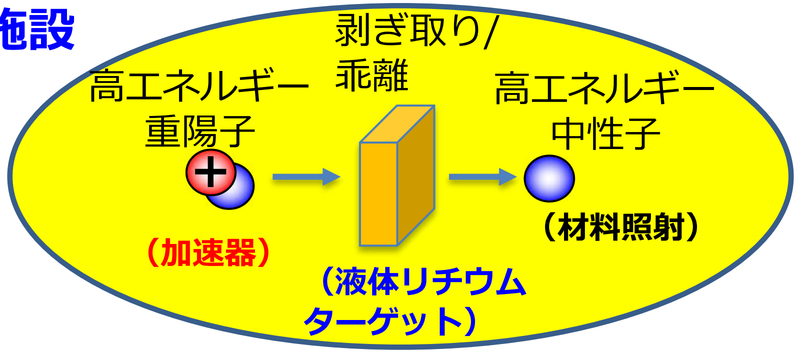
QST

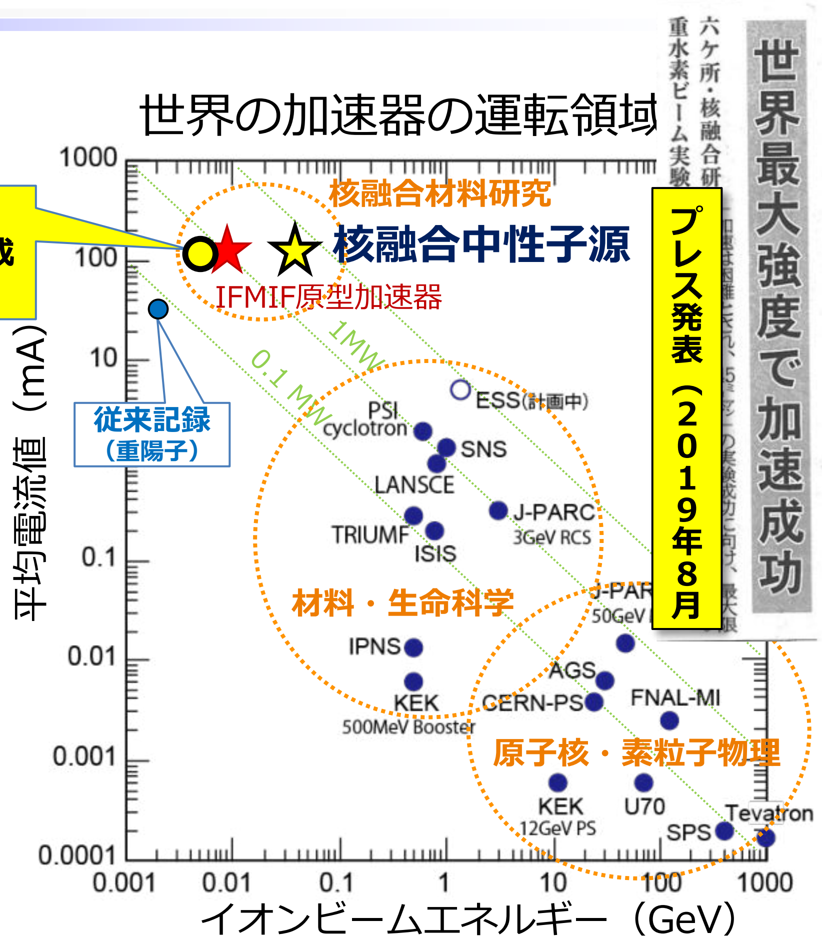
核融合炉では、多量の高速中性子が周辺壁に飛来。

課題
発生エネルギーを取出す
（中性子を受止め、熱に変える）



核融合中性子源**：加速器を用いて、核融合炉と同様の大量の高速中性子を発生させて材料の耐久性評価を行う施設**





世界に類のない大電流加速器(125mA、40MeV):右図

⇒ 技術基盤となる**IFMIF原型加速器(125mA/9MeV)を六ヶ所研において日欧開発中**

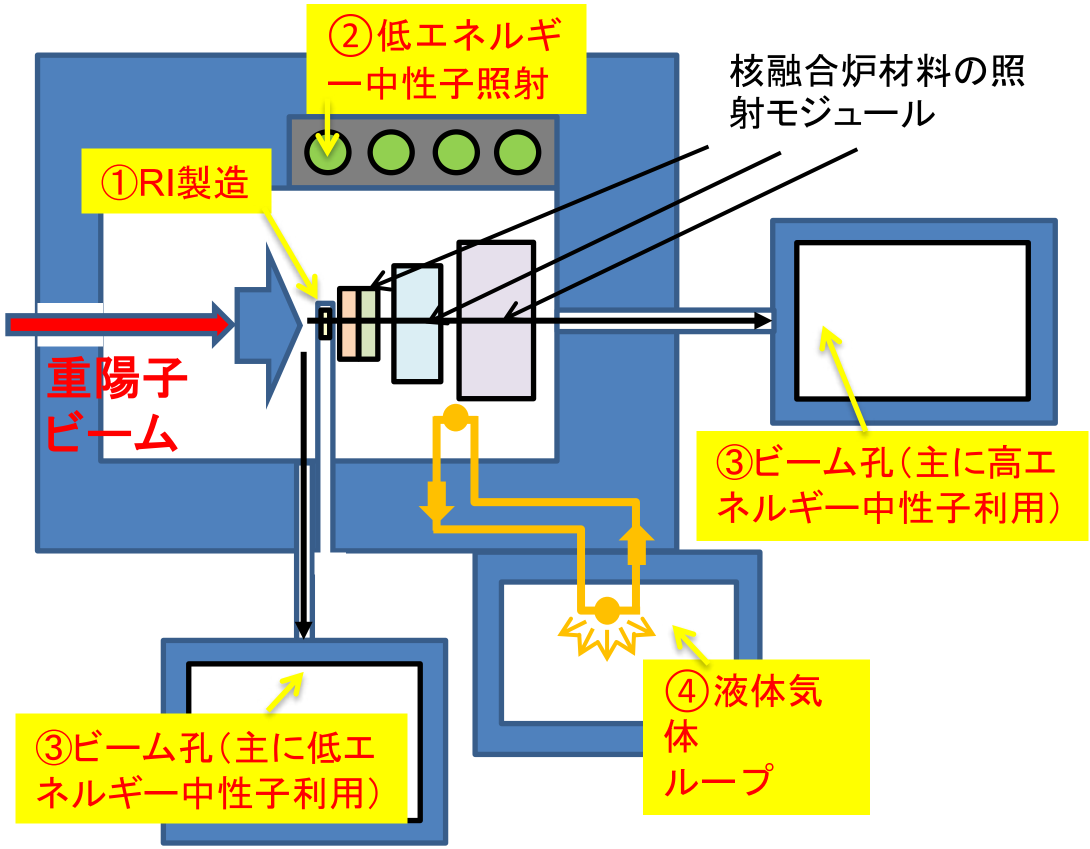
⇒ 2019年7月、電流として**世界最高値125mAを５MeV、短パルスで達成**

⇒ 今後、125mA/5MeVの長パルス試験の後、 125mA/９MeVの実証を目指す

その成果を基に、核融合中性子源の建設を目指す

A-FNSにおける応用利用の概要

核融合炉の材料や機器試験だけでなく、基礎研究・産業や医療・エネルギー応用への展開を図る　⇒　A-FNSで実施する応用利用を4つに整理



①多目的RI生成モジュール

- 医療用RI製造（Mo-99など）⇒*Tc-99
- 箔放射化法による中性子束計測

②低エネルギー中性子照射モジュール

- シリコン半導体製造
- 低エネルギー中性子照射

③多目的利用ビーム孔システム

- 中性子イメージングによる高速動画撮影
- 核データ研究

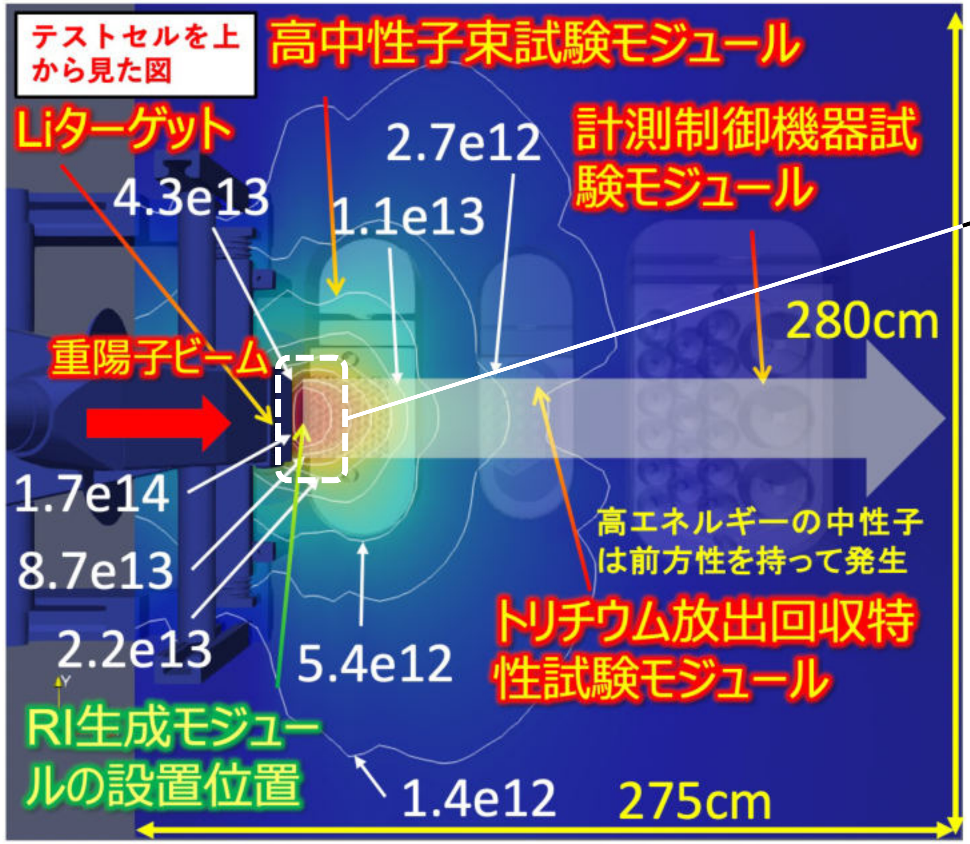
④液体気体ループ照射試験モジュール

- ガンマ線や陽電子を用いた分析や照射
- 水の放射化法を用いた中性子モニター

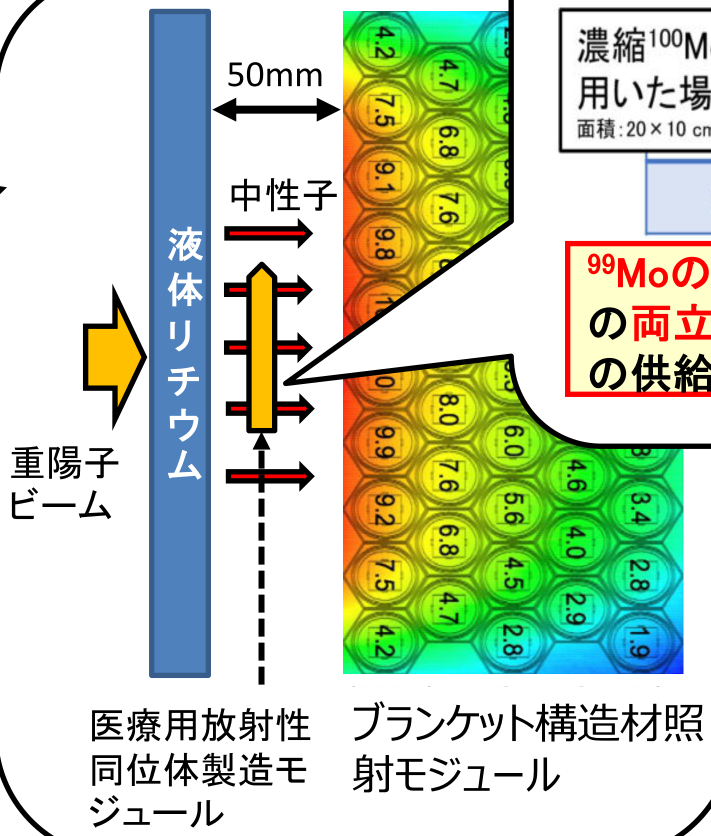
中性子束、照射期間などの観点から、核融合炉材料の照射試験に影響を与えない中性子利用

中性子照射の産業応用の例（RI製造モジュール）

モジュールがある場合の中性子束



拡大図



日本の需要を満たす^{99}Mo製造が可能

濃縮$^{100}MoO_3$試料を用いた場合（5ミリ厚） 面積：20×10 cm^2, Net ^{100}Mo：317 g	放射能（TBq/回）と日本の需要※に対する割合 [%]
3 日照射	83 [99%]

^{99}Moの大量製造と、核融合炉材料照射試験の両立が可能、原子炉を用いなくても、^{99}Moの供給が可能

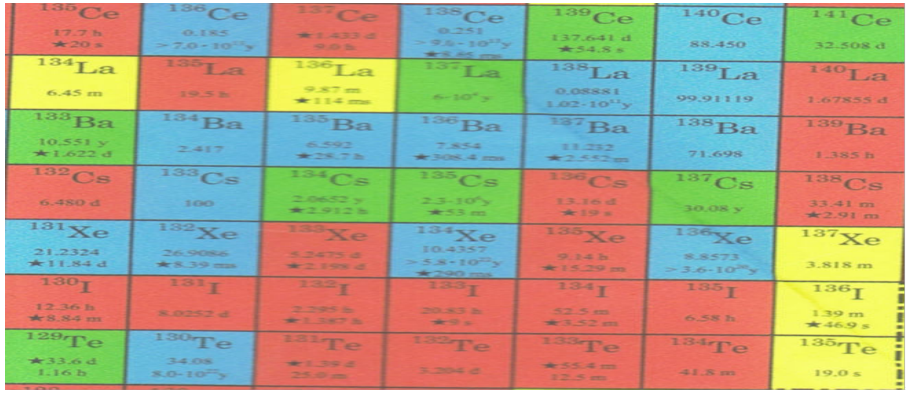
医療用放射性同位体製造モジュールを挿入することによる影響はわずか1%と評価

核融合炉材料用照射と医療用放射性同位体製造の概念を構築し、同時に利用できるように最適化することにより、核融合材料照射と中性子を用いた医療用放射性薬剤製造の両立に展望を拓いた。

IV. 超長半減期FPの核変換処理の可能性

- Application of IFMIF on the transmutation processing of the long life fission products of **^{79}Se, ^{93}Zr, ^{99}Tc, ^{107}Pd,^{126}Sn, ^{129}I,^{135}Cs** from nuclear fuel.
- Processing of these FPs processed in fission reactors without isotope separation, **(n,γ)reaction** takes place with high cross sections of low energy neutrons. This produces LLFP from stable or short half life elements, so that the complete elimination of LLFP is difficult with using fission reactors.
- With high energy neutrons this is adverse changed owing to lower cross section of (n,γ) to those of **(n,2n), (n,3n), (n,p), (n,np)** in these energy region.

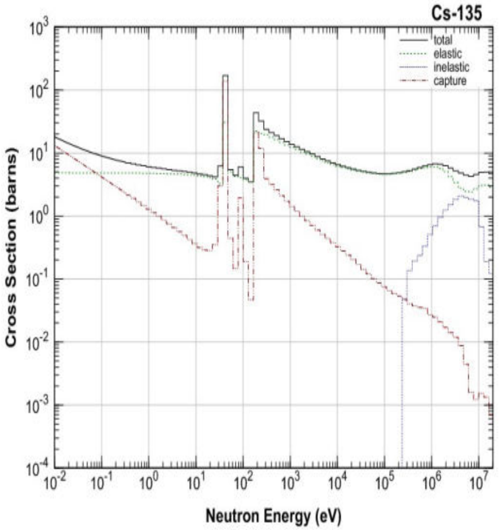
断面積を詳細に調べる必要がある



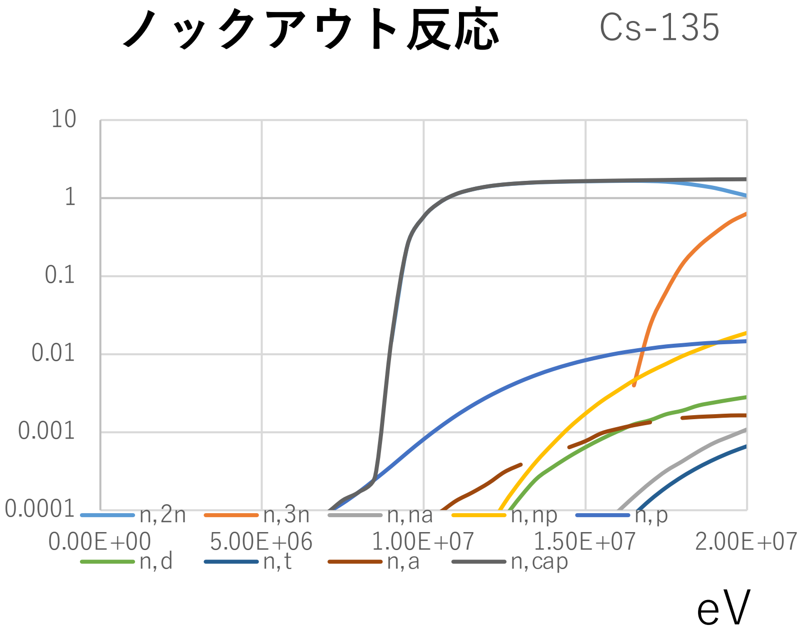
Thermal neutrons enhance the neutron capture reactions (n,γ). Long life elements are produced from short life FP elements.

$^{133}Cs \Rightarrow ^{134}Cs \Rightarrow ^{135}Cs \Rightarrow ^{136}Cs \Rightarrow ^{137}Cs$

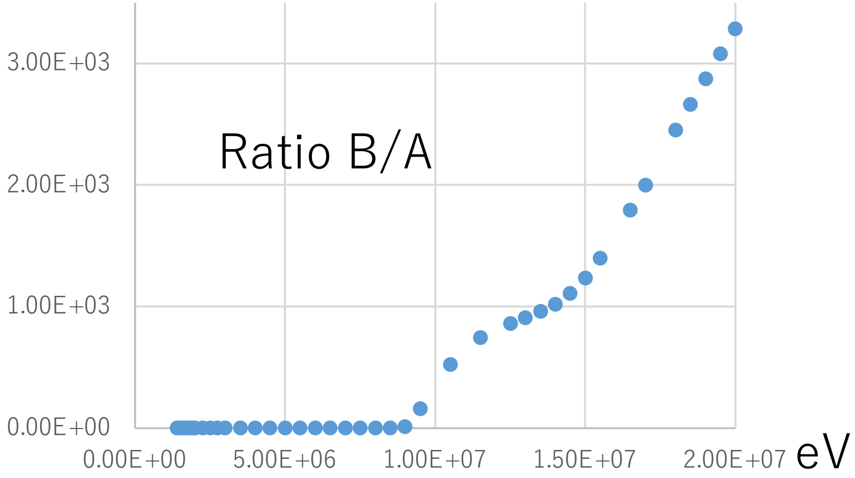
A:(n,γ)中性子捕捉



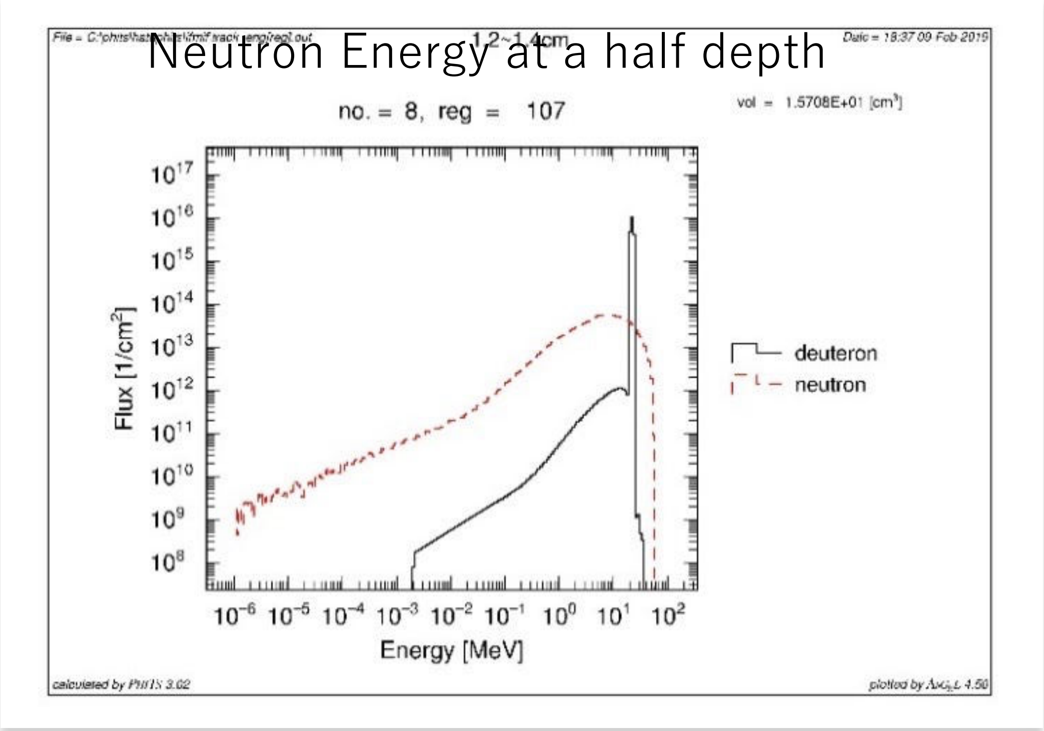
B:(n,2n),(n,t), (n,p)---
ノックアウト反応



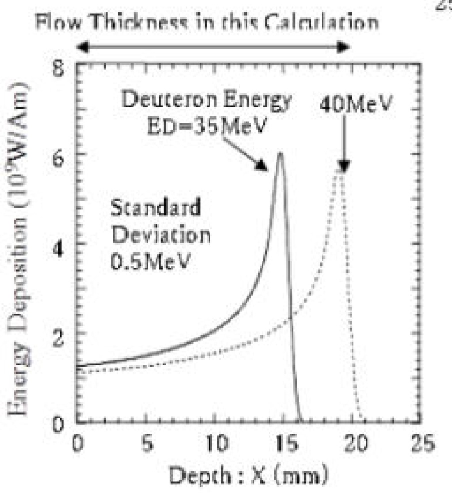
20-30MeV neutrons raise (n,2n) like reactions which is 2-3 orders higher than (n,γ).

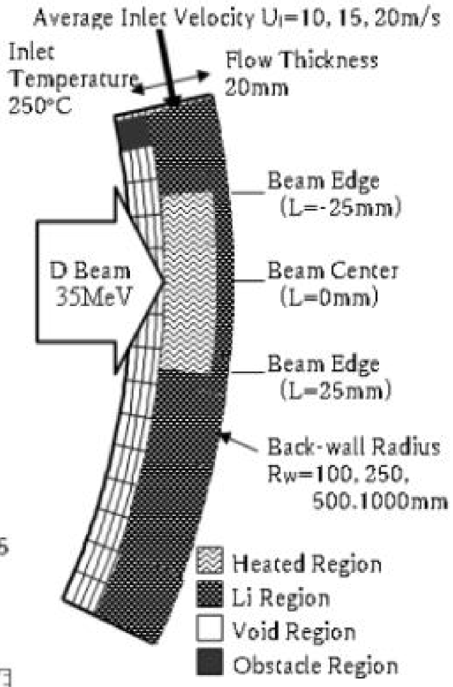


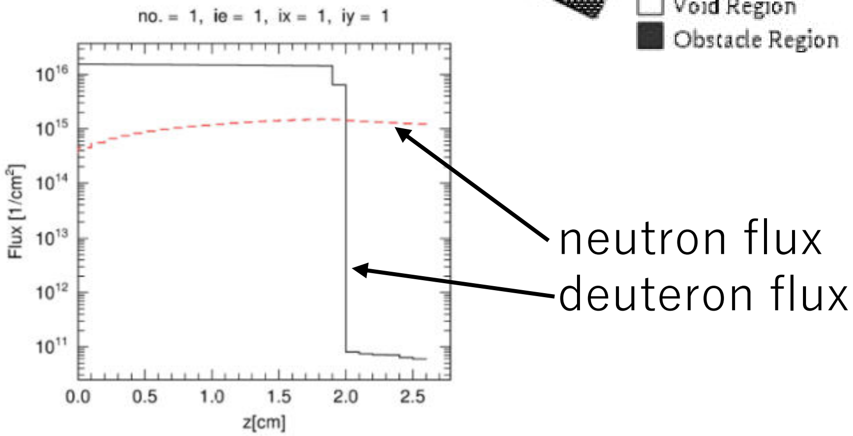
Transmutation system in Li flow



Li is light so that the neutron moderation is very small within the depth of Li.
LLFP's could be mixed into Li fluid to be irradiated continuously by 10-40MeV neutrons, avoiding (n,γ)reaction.

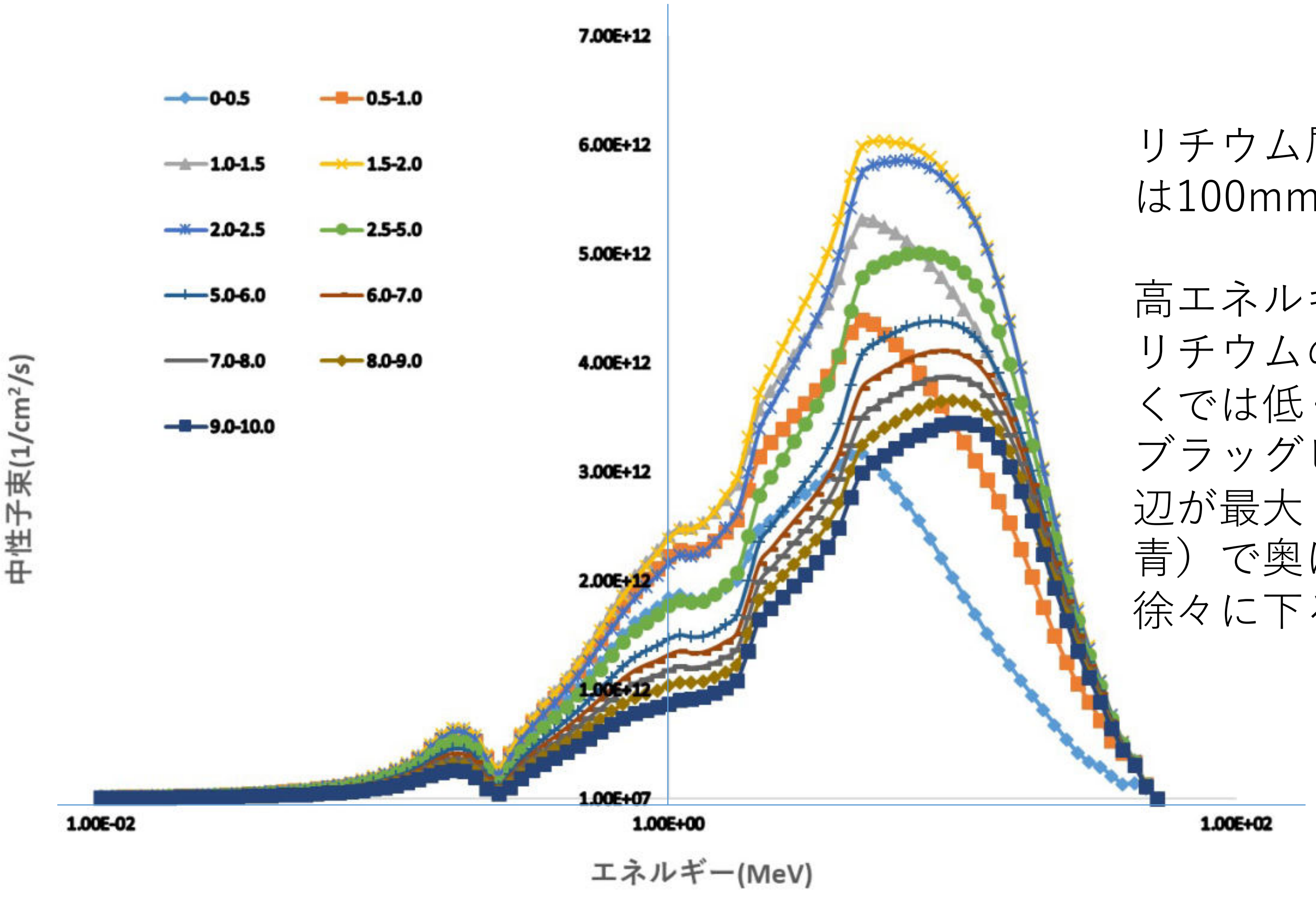






Processing at high rate requires large beam current

Li内の中性子束の強度分布

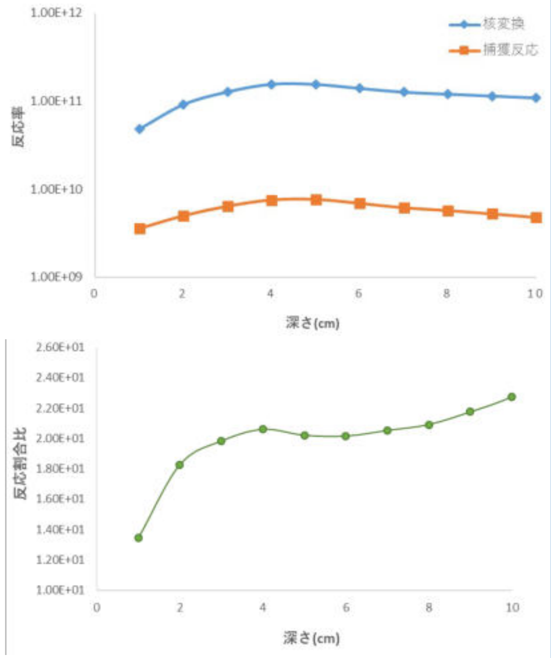


リチウム層の厚さは100mmを仮定

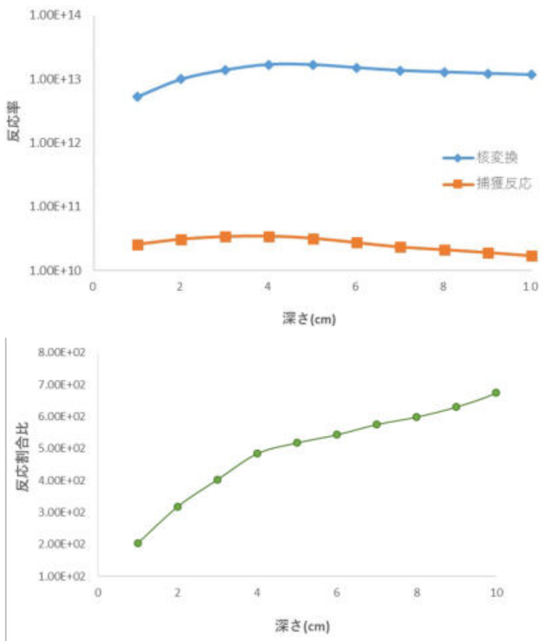
高エネルギー束はリチウムの表面近くでは低く（青）、ブラッグピーク周辺が最大（橙、青）で奥に向けて徐々に下る

核変換(青）と捕獲（橙）の比（緑）

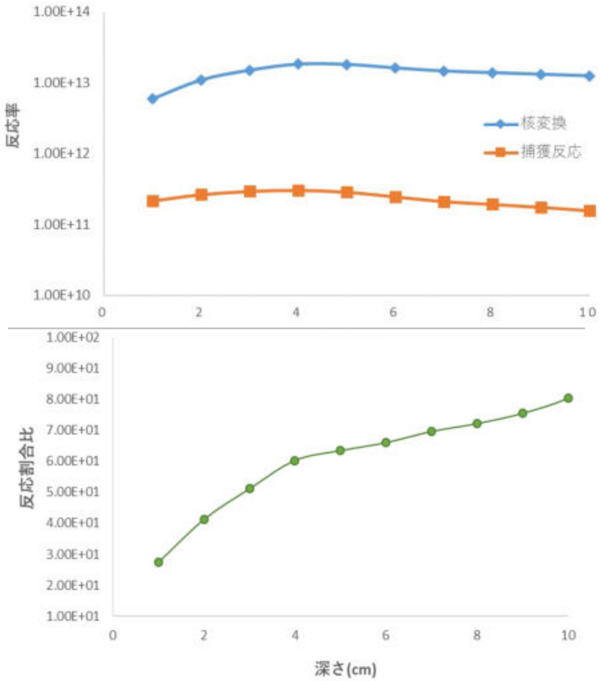
Se ~20



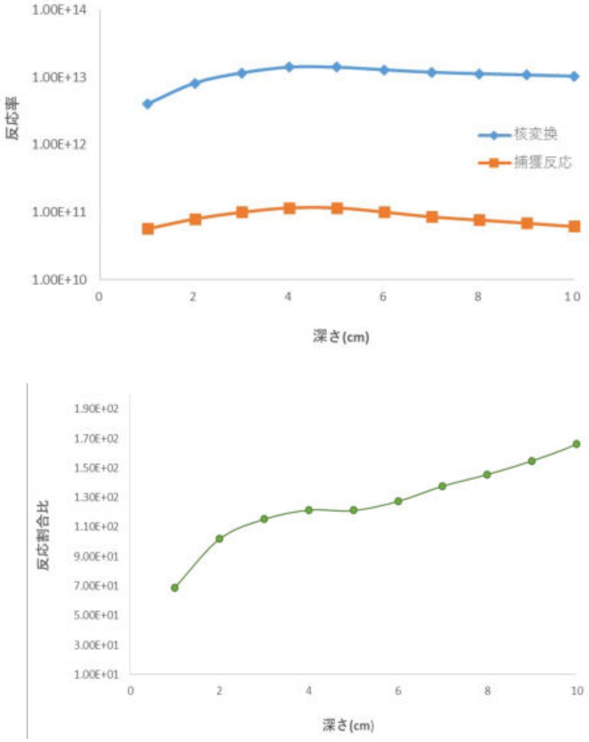
Zr ~70



Pd ~80



Cs ~170



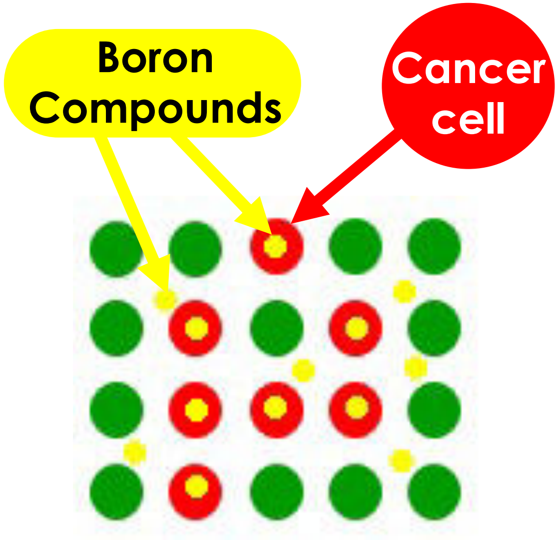
核種	質量(g)
^{79}Se	5.5541
^{93}Zr	678.34
^{107}Pd	683.85
^{135}Cs	1017.7

核変換の可能性のまとめ

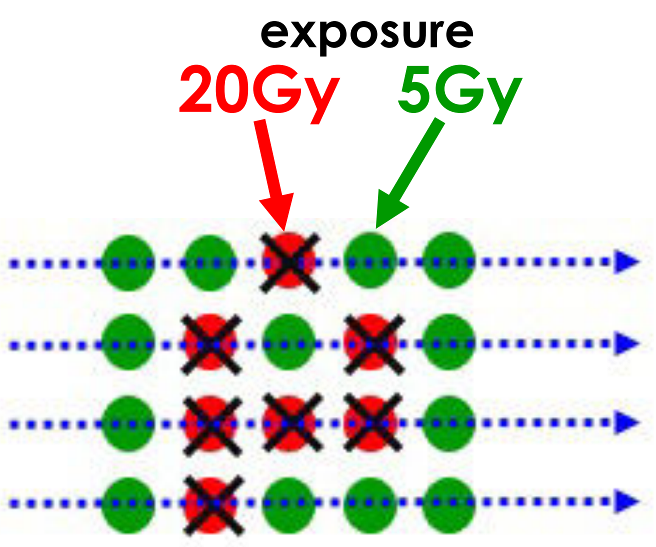
- 10 to 40MeVの中性子： $\sigma_{(n,\gamma)}$ ＜ $\underline{10^{-3}\ \text{barn},}$
  ： $\sigma_{knockout}$ ＞ $\underline{1\ \text{barn}}$
- Li流の後方において約100倍の変換比が可能
- この数字は解析値でありA-FNSにより精確に測る価値がある
- 20~30年後の消滅処理の実用化が期待できる
- 実用化には高エネルギーで大電流の加速器が必要

V. Boron Neutron Capture Therapy

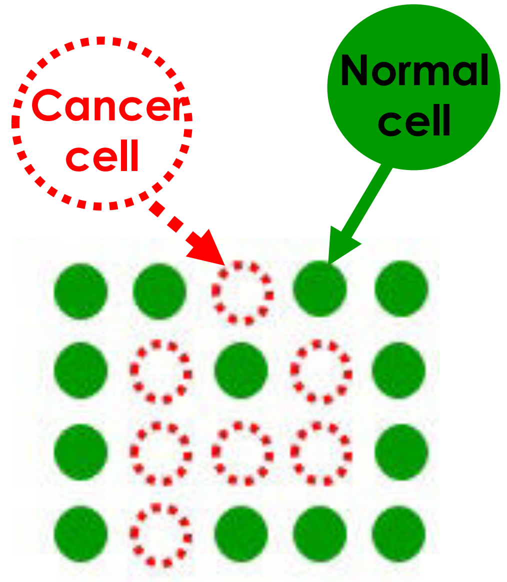
BNCT is a kind of particle beam therapy that selectively destroys cancer cells with using reaction between a slow neutron and a boron accumulated in a cancer cell.



Nutrient with boron is swallowed by active cancer cells.



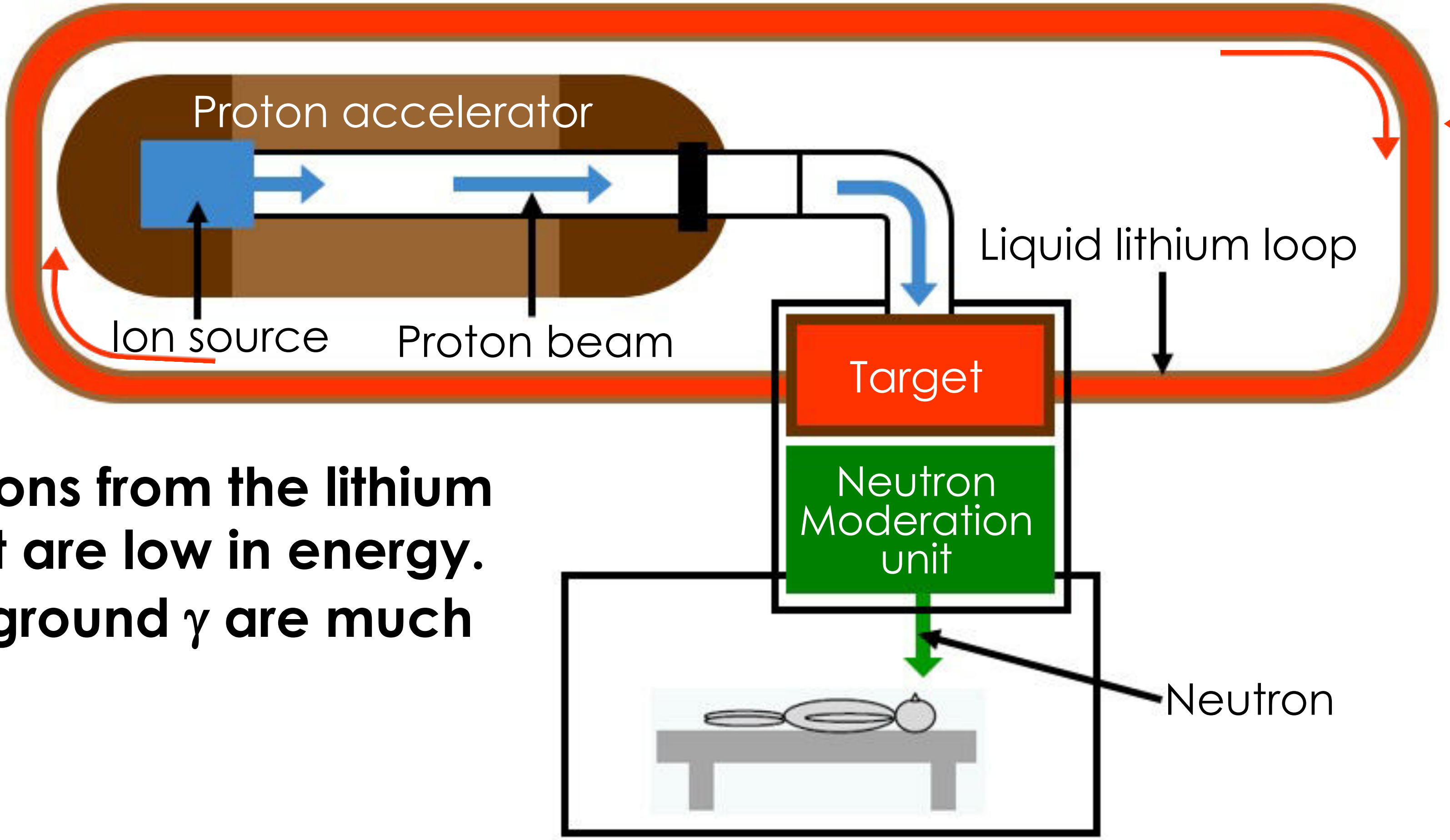
$^{10}B + n \rightarrow$ He + Li + 2.31MeV + γ ray
where
E_{α}=1.47MeV, E_{Li}=0.84MeV, E_{γ}=478 keV
Ion's ranges < 5 – 7mm < cell diameter



Energetic ions work as a heavy ion therapy. Cancer cells with many boron are selectively destroyed.

<Source: Association For Nuclear Technology in Medicine website>

Liquid Lithium Neutron Source



Liquid lithium target

- **Low exposure**
- **Stable operation**
- **No replacement of target**

Neutrons from the lithium target are low in energy. Background γ are much less.

Experiments on neutronics completed

Li neutron production test in Birmingham University

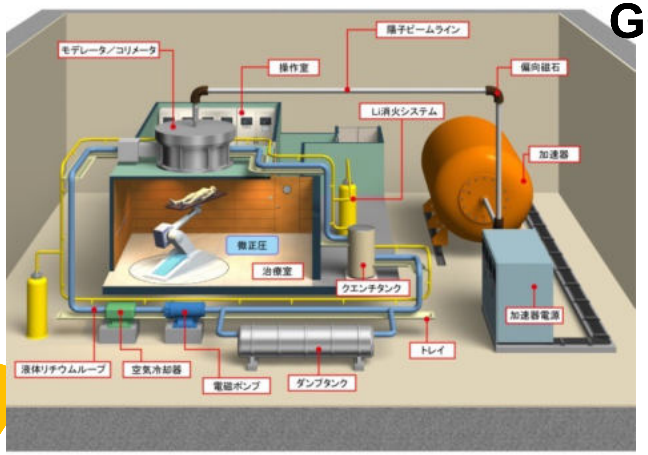
2012
Tohoku Univ.

Source term exp.

2013
Birmingham Univ

Moderator assembly exp.

GOAL



Design Value : 30mA 2.5MeV

Neutron Flux: **10^9/sec・cm^2**

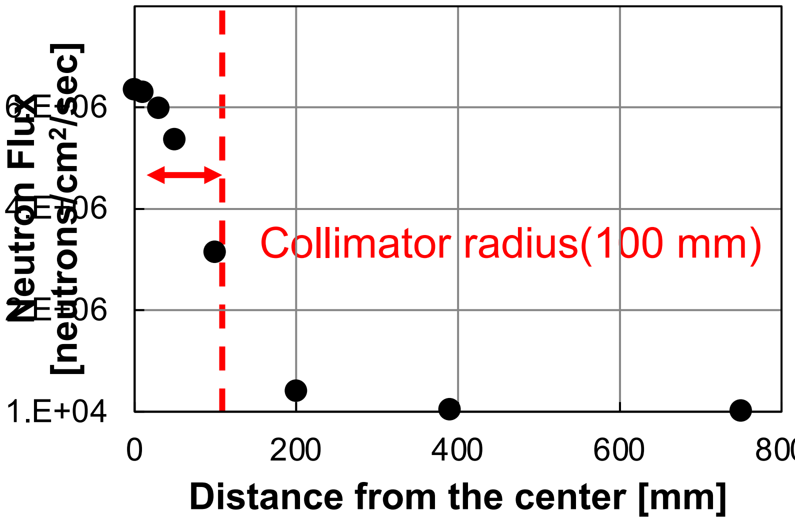
1μA — 1mA — 30mA Ampere

1. Produced neutron properties and accompanying gamma rays and high energy components of neutrons were studied in detail.
2. Calibration of numerical code system, and verification of irradiation performances up to 30mA.

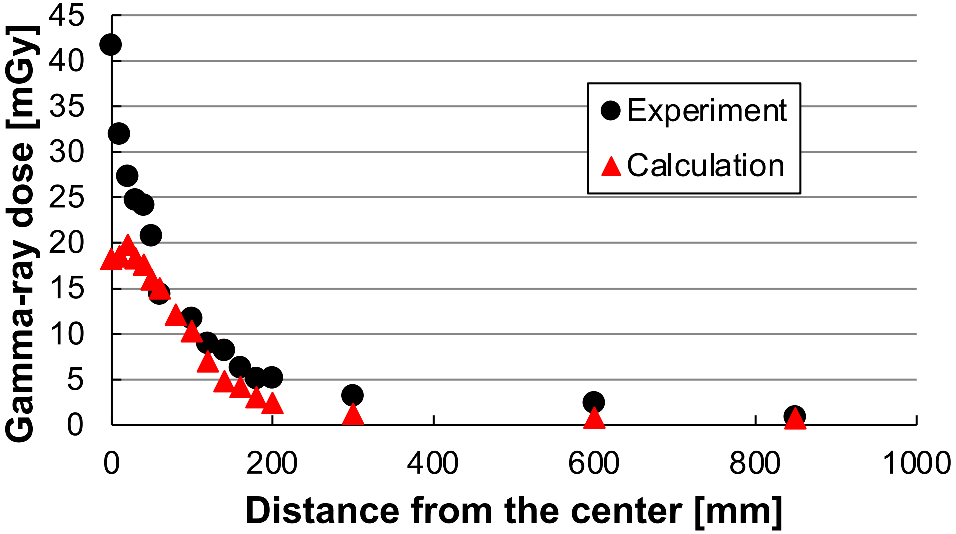
Neutron Flux Measurements

2.65 MeV w/o phantom

Epithermal neutron profile



Gamma ray profile

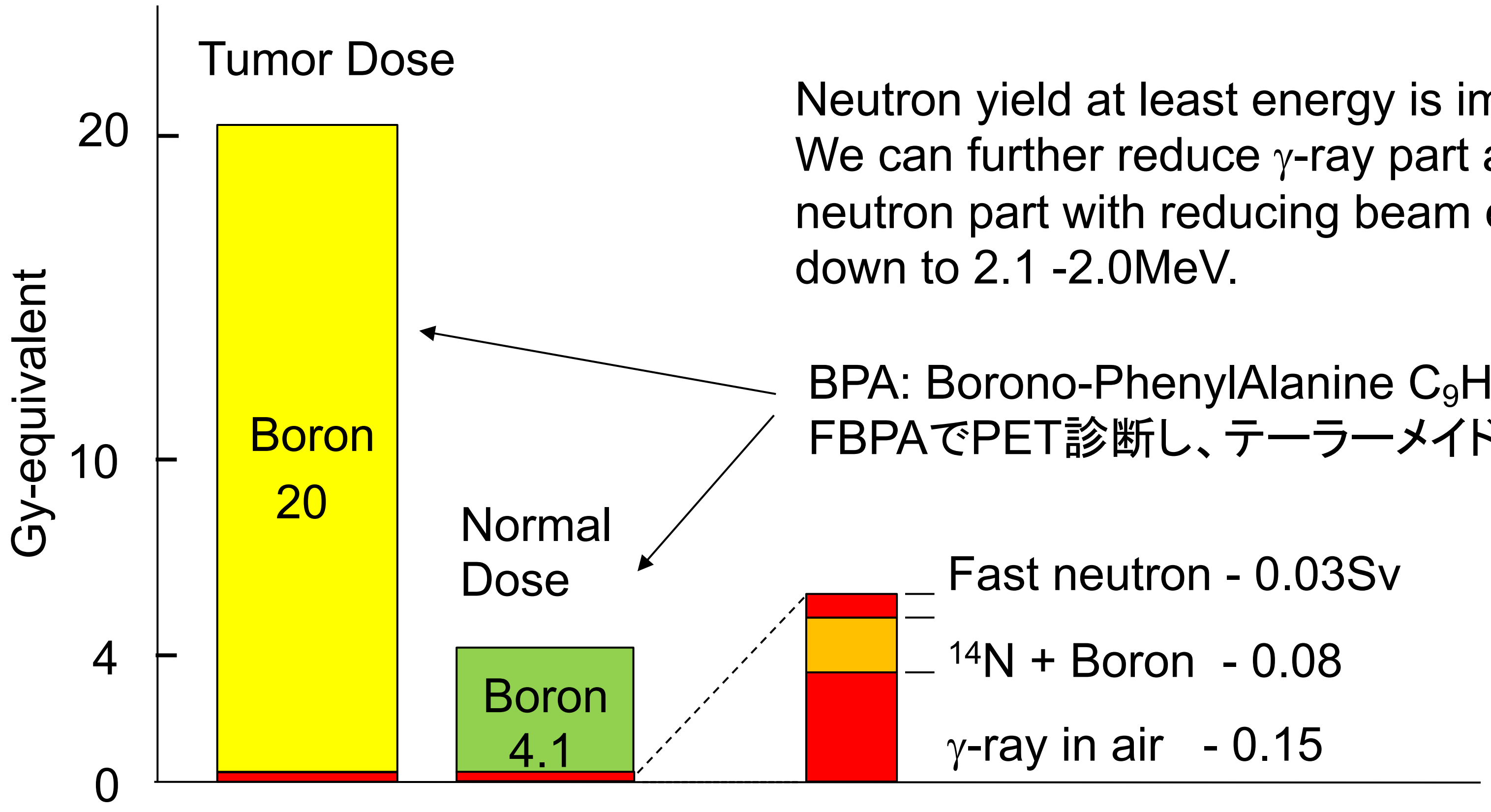


- **Neutron flux is collimated well and low beyond collimated radius.**
  → Suppression of the total body dose for a patient
  Shingo Tamaki, "Mock-up Experiment at Birmingham University for BNCT Project of Osaka University - Neutron Flux Measurement with Gold Foil –" ***Poster Sessions (Ps2 P01) ICNCT2014 June Helsinki 2014***
- **A peak of the gamma at the center is partly attributable to neutron doses, which has verified by numerical analysis and by new differential measurement of γ-ray.**
  → A glass dosimeter is tested to have sensitivity to neutrons.
  Sachiko Yoshihashi, "Mock-up Experiment at Birmingham University for BNCT Project of Osaka University - Gamma-ray Dose Measurement with Glass Dosimeter – " ***Poster Sessions (Ps2 P03) ICNCT2014 June Helsinki 2014***

Real Machine Performance 1

Neutron yield at least energy is important. We can further reduce γ-ray part and fast neutron part with reducing beam energy down to 2.1 -2.0MeV.

BPA: Borono-PhenylAlanine $C_9H_{12}BNO_4$
FBPAでPET診断し、テーラーメイド医療に

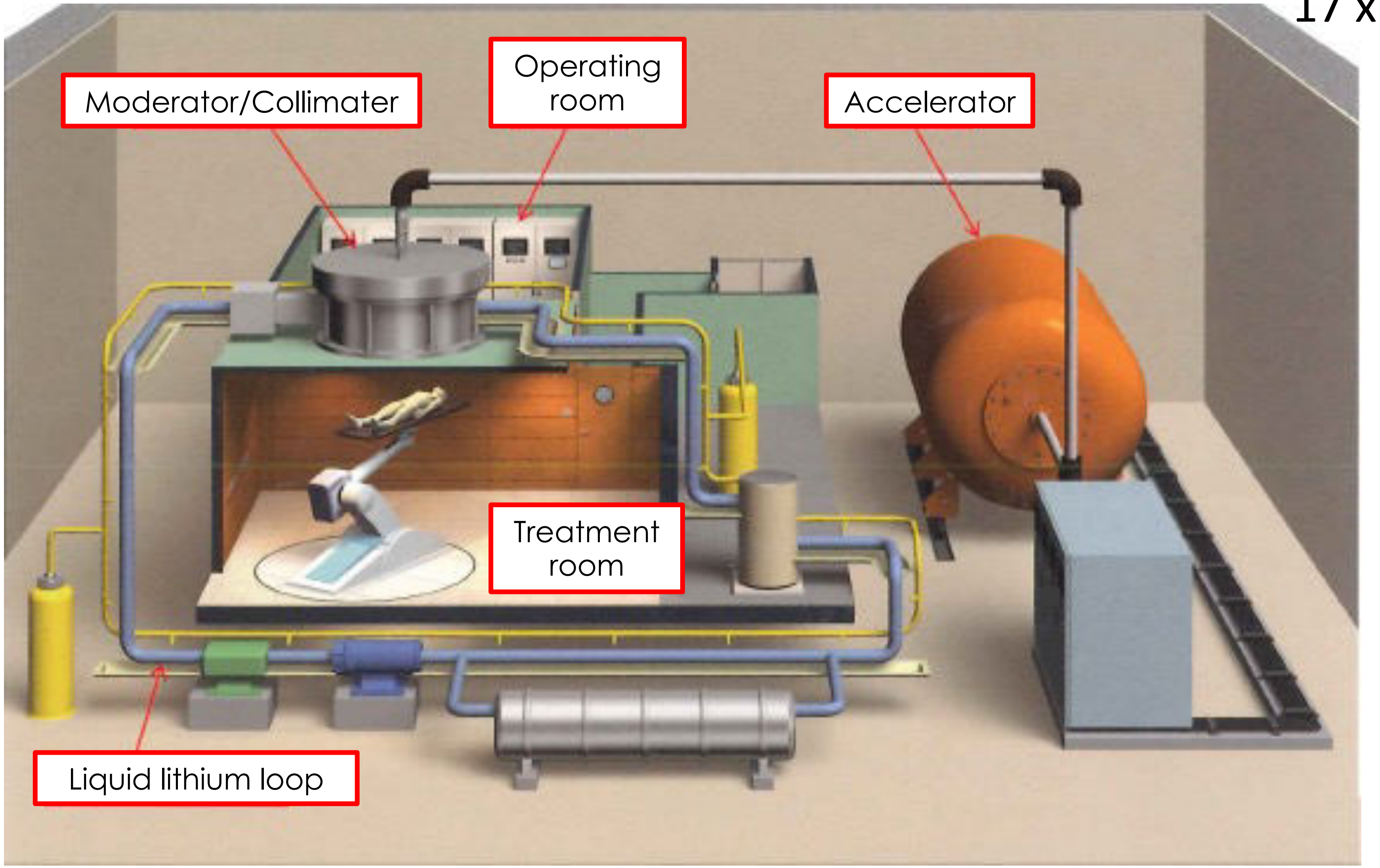


(TOTAL = 0.26 Sv/treatment)

安全且つ簡便で、良く治るがん治療が可能

Estimated completion of the system

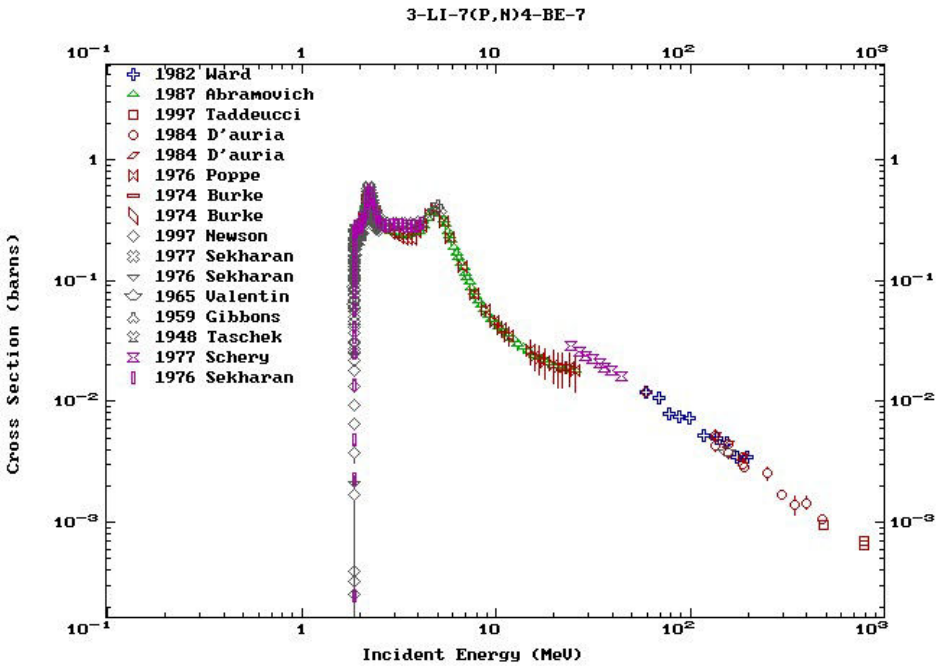
$17 \times 17 \times 5 m^3$



VI. About Accelerator

Above described application requires,

- 30-60mA at 2.5MeV H+ for BNCT,
- 1A at 40MeV D+ for transmutation.



- RFQ has limitations
- JT-60SA 20A 500kV D^-
- ITER 40A 1000kV D^-
- With using these high voltage knowledge, **<u>high current tandem accelerator</u>** at 2500kV is fabricated.

CONCLUSION

- Liquid Lithium circulation technology has advanced owing to the series of experimental projects.
- Contraction of the flow with two stage nozzle was validated to perform satisfactorily for a high speed free surface flow.
- Stable free surface flow of up to 20m/sec under 10^{-3}Pa vacuum, its long time stability are verified with spindle and laser probes and with numerical analysis.
- Various application of Li flow combined with accelerator are derived, e.g. transmutation of LLFP, BNCT and FRIB stripper.
- High current MeV beams are desirable, 100mA for BNCT and 1A for LLFP disposal.
- These current region may easily be satisfied with a tandem accelerator with using the MeV-class ion source technology and high voltage power supply of current SiC semiconductors.