医療用薬剤$^{99}Tc^m$の製造：γ核反応

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RCNP研究会「RCNP加速器増強と核破砕破砕反応中性子利用」
@RCNP, 2012/9/28-29

本発表の内容

○医療用薬剤$^{99}Tc^{m}$の製造：γ核反応

- $^{99}Mo/^{99}Tc^{m}$製造の現状

- $^{99}Mo/^{99}Tc^{m}$の安定供給に向けて
（原子炉以外の$^{99}Mo/^{99}Tc^{m}$製造法）

- （現有の）加速器による$^{99}Mo/^{99}Tc^{m}$製造の見積もり
  - プロトン照射
  - 制動放射線照射

- まとめ

$^{99}Mo/^{99}Tc^m$について

○$^{99}Tc^m$：核医学検査の中で利用件数が最も多い
放射性核種 （年間90万件）

○^{99}Mo：$^{99}Tc^m$の親核種。日本は100%輸入

○^{99}Moは高濃縮ウランを用いて原子炉（研究炉）
で製造。世界で5-6か所

世界的な^{99}Moの供給不足

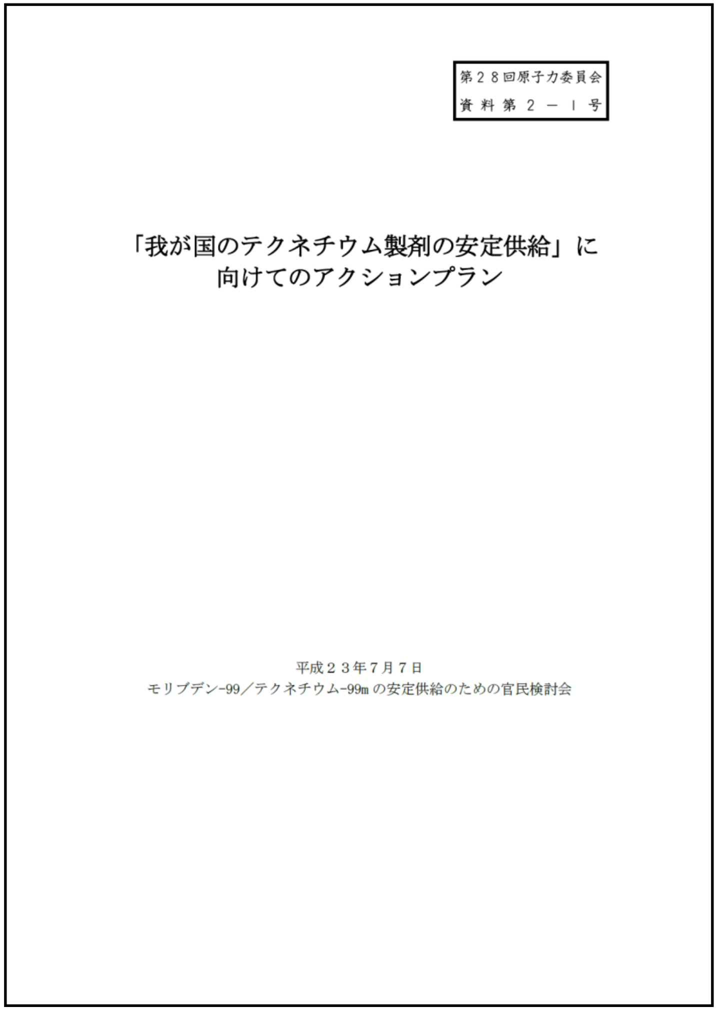
2009- ^{99}Mo製造用原子炉のトラブル
2010 火山噴火により^{99}Moの航空輸送ストップ

⇒ 病院での検査に支障

○原子炉の老朽化により世界的な^{99}Mo供給不足の可能性

⇒ 各国で安定供給の方策を議論

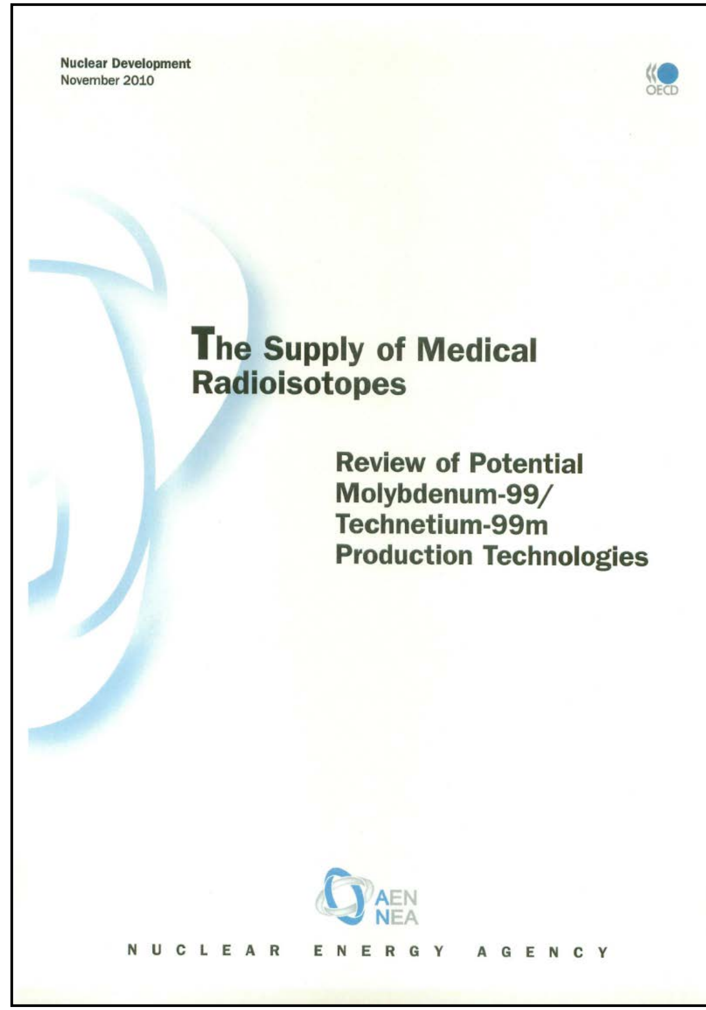
^{99}Mo/^{99}Tc^{m}の安定供給に向けて


第２８回原子力委員会
資料第２－１号

「我が国のテクネチウム製剤の安定供給」に
向けてのアクションプラン

平成２３年７月７日
モリブデン-99／テクネチウム-99m の安定供給のための官民検討会

Japanese report, 2012


Nuclear Development
November 2010

OECD

The Supply of Medical Radioisotopes

Review of Potential Molybdenum-99/ Technetium-99m Production Technologies

AEN NEA

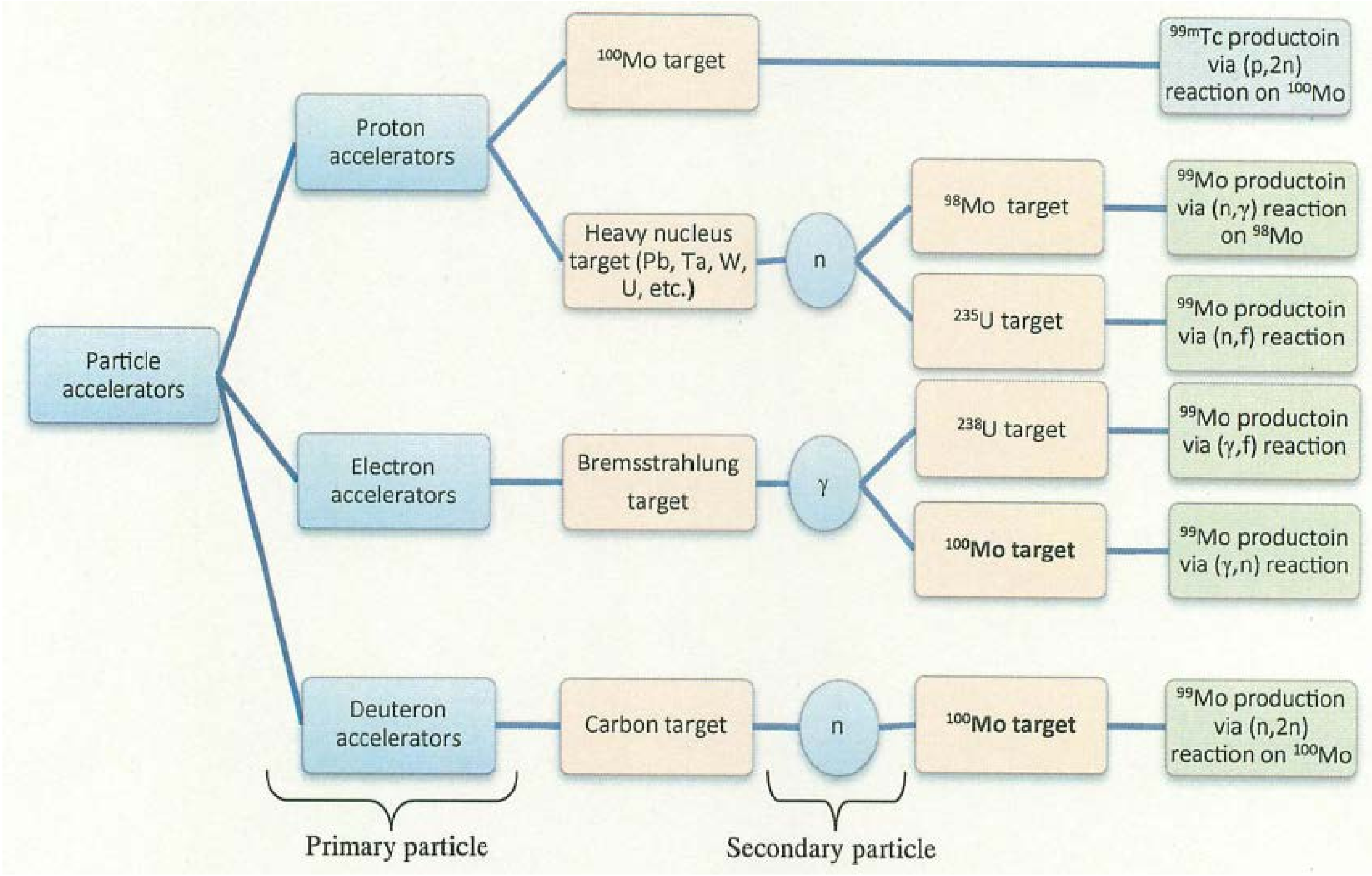
NUCLEAR ENERGY AGENCY

OECD/NEA report, 2010

安定供給の方策の一つが国産^{99}Mo

加速器を用いた^{99}Mo製造

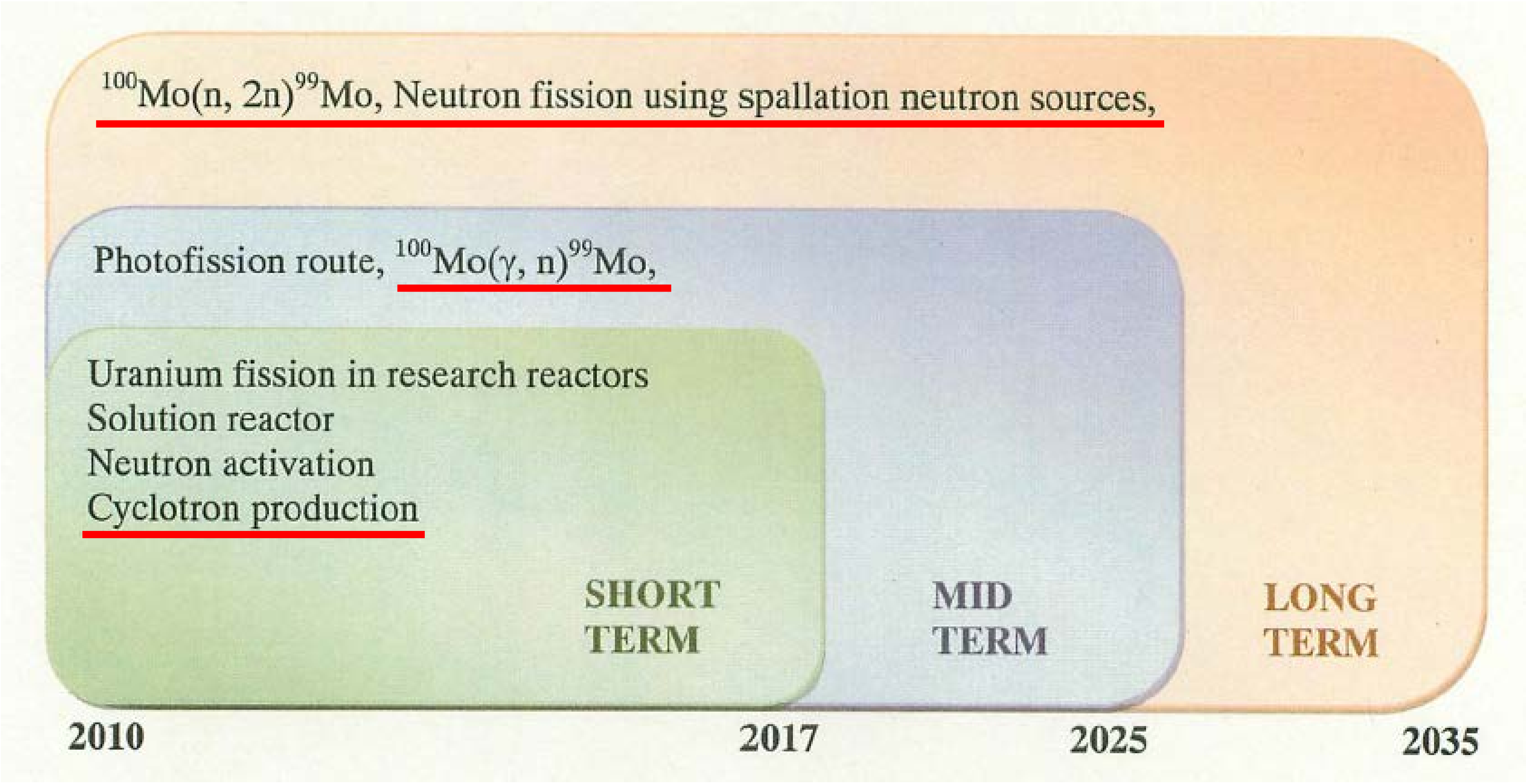
〇核拡散等の問題から原子炉を用いない^{99}Mo製造が提案されている。



Summary of potential accelerator-based ^{99}Mo/^{99m}Tc production technologies

OECD/NEA report, 2010

^{99}Mo安定供給計画



Classification of technologies used in the report

OECD/NEA report, 2010

プロトンビームを用いた^{99}Mo製造

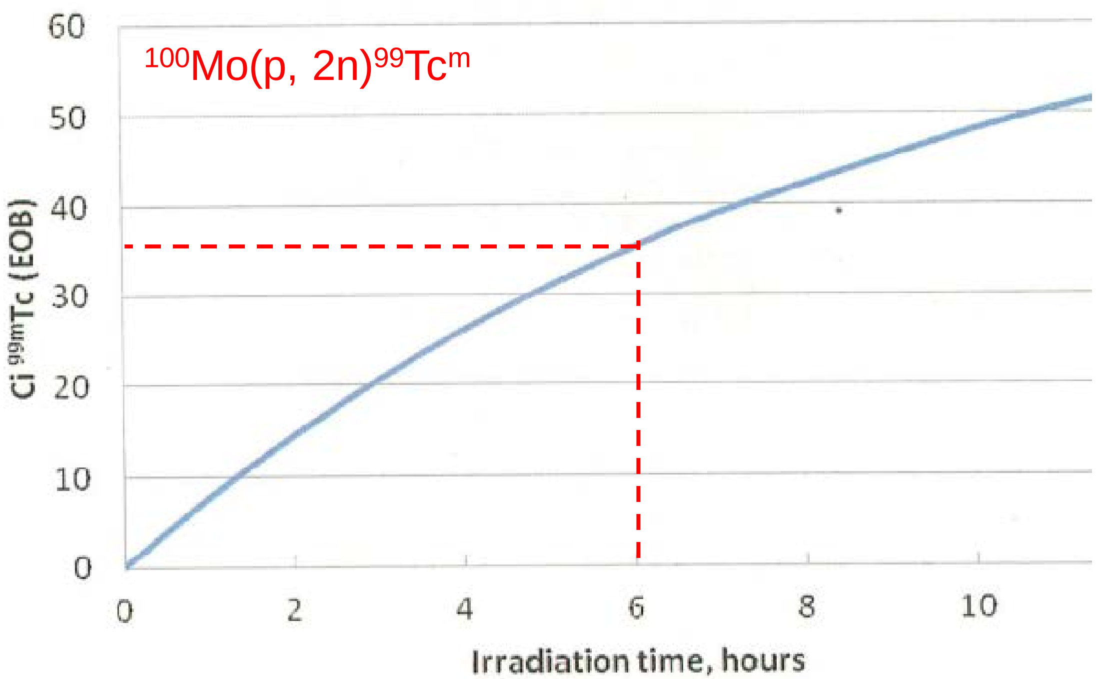
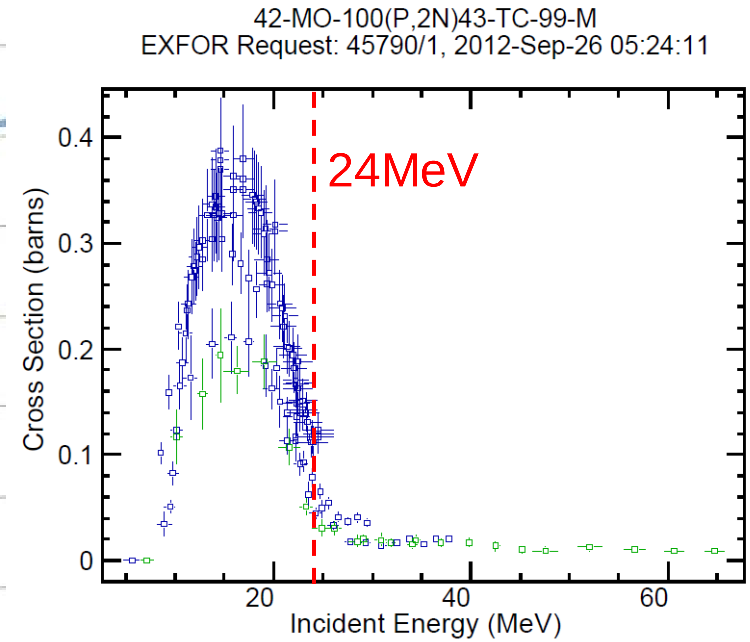


Figure 10: Direct ^{99m}Tc production in a 500 μA cyclotron at 24 MeV

Enriched ^{100}Mo metal target (~1000 mg/cm^2)



〇10-30 μA，6 h照射で700-2100 mCi（30-90回の検査；25 mCi/1回）

OECD/NEA report, 2010

海外でのRI製造

Session 7 - Nuclear Chemistry, Radionuclide Production, High-Power Targetry - (08:00-10:45)

- Conveners: Prof. Zhuikov, Boris (Institute for Nuclear Research of Russian Academy of Sciences, Russia); Prof. Bonardi, Mauro L. (UNIMI and INFN - Milano, Italy)

Session 8 - Nuclear Chemistry, Radionuclide Production, High-Power Targetry - (11:00-13:30)

- Conveners: Prof. Srivastava, Suresh (Brookhaven National Laboratory, USA); Dr. Mikolajczak, Renata (Polatom, Poland)

2/13セッションがRI製造

time	[id] title	presenter
11:00	[245] OPENING LECTURE - The Road to Cyclotron Produced Tc-99m	Prof. MCQUARRIE, Steve (University of Alberta, Canada)
11:20	[32] INVITED LECTURE - Cyclotron production of radionuclides with medium-energy proton beams and high-power targetry	Dr. STEYN, G. (iThemba LABS, South Africa)
11:40	[29] INVITED LECTURE - Radionuclide Production at Accelerator with High Power Targets	Dr. ZHUIKOV, Boris (Institute for Nuclear Research of Russian Academy of Sciences, Russia)
12:00	[242] INVITED LECTURE - Recent advances in large scale isotope production at LANL	Dr. NORTIER, Francois (Los Alamos National Laboratory, USA)
12:20	[237] INVITED LECTURE - ARRONAX: on the way to the production of radio-isotopes with high-power targets	Dr. HADDAD, Ferid (Subatech / ARRONAX, France)
12:40	[238] INVITED LECTURE - Use of radioactive targets for production of therapy radionuclides at the Brookhaven Linac Isotope Producer	Dr. MAUSNER, Leonard (Brookhaven National Laboratory, USA)

EuCheMS International Conference on Nuclear and Radiochemistry (NRC-8)
Como, Italy, Sep.16-21 (2012) 講演プログラムより

海外でのRI製造

12:20	[237] INVITED LECTURE - ARRONAX: on the way to the production of radio-isotopes with high-power targets	Dr. HADDAD, Ferid (Subatech / ARRONAX, France)

ARRONAX is unique through the association of several characteristics:
-a 70 MeV energy whereas the energy of the majority of biomedical cyclotrons
-does not exceed 30 MeV - a maximal intensity of 750 µA (2 X 3755 µA)whereas
-the intensity of the majority of biomedical cyclotrons does not exceed 100 µA
-- the possibility to accelerate protons, deutons and alpha particles whereas
-the majority of biomedical cyclotrons accelerates only protons.

ARRONAX HPより

〇（NRC8の発表では）いくつかの施設で300-1500 µAでRI製造

プロトンビームを用いた^{99}Mo製造

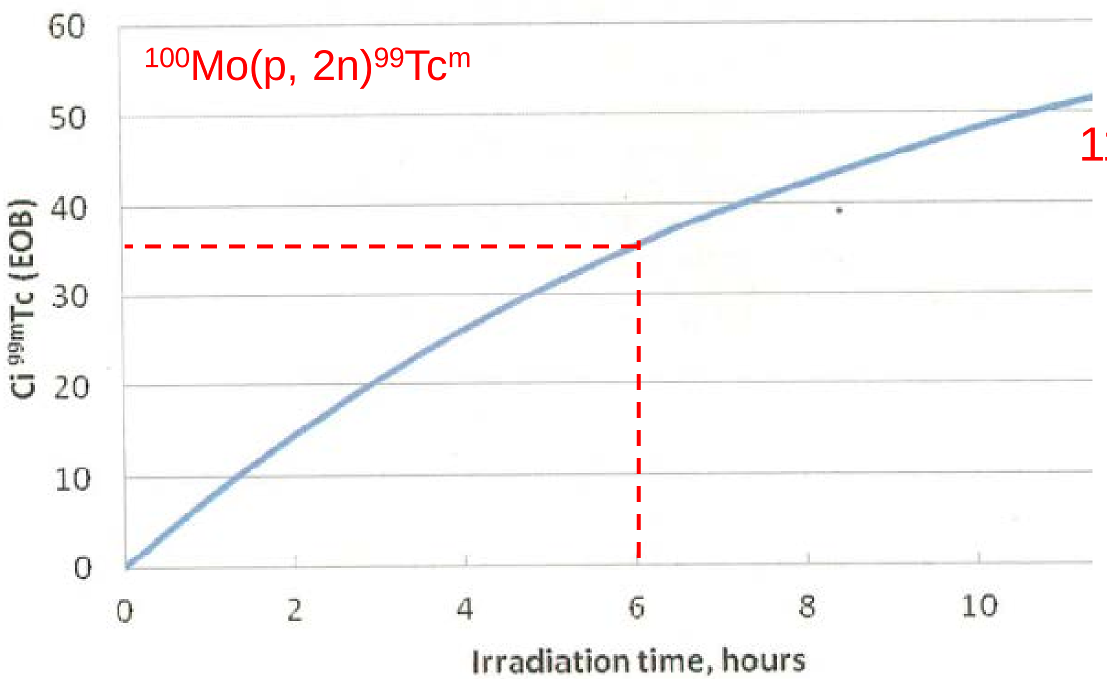
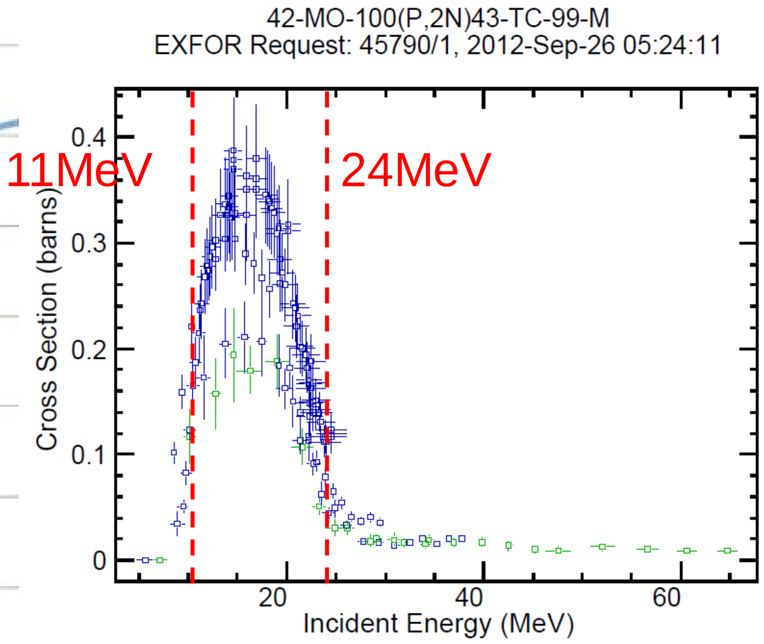


Figure 10: Direct ^{99m}Tc production in a 500 μA cyclotron at 24 MeV

Enriched ^{100}Mo metal target (~1000 mg/cm^2)

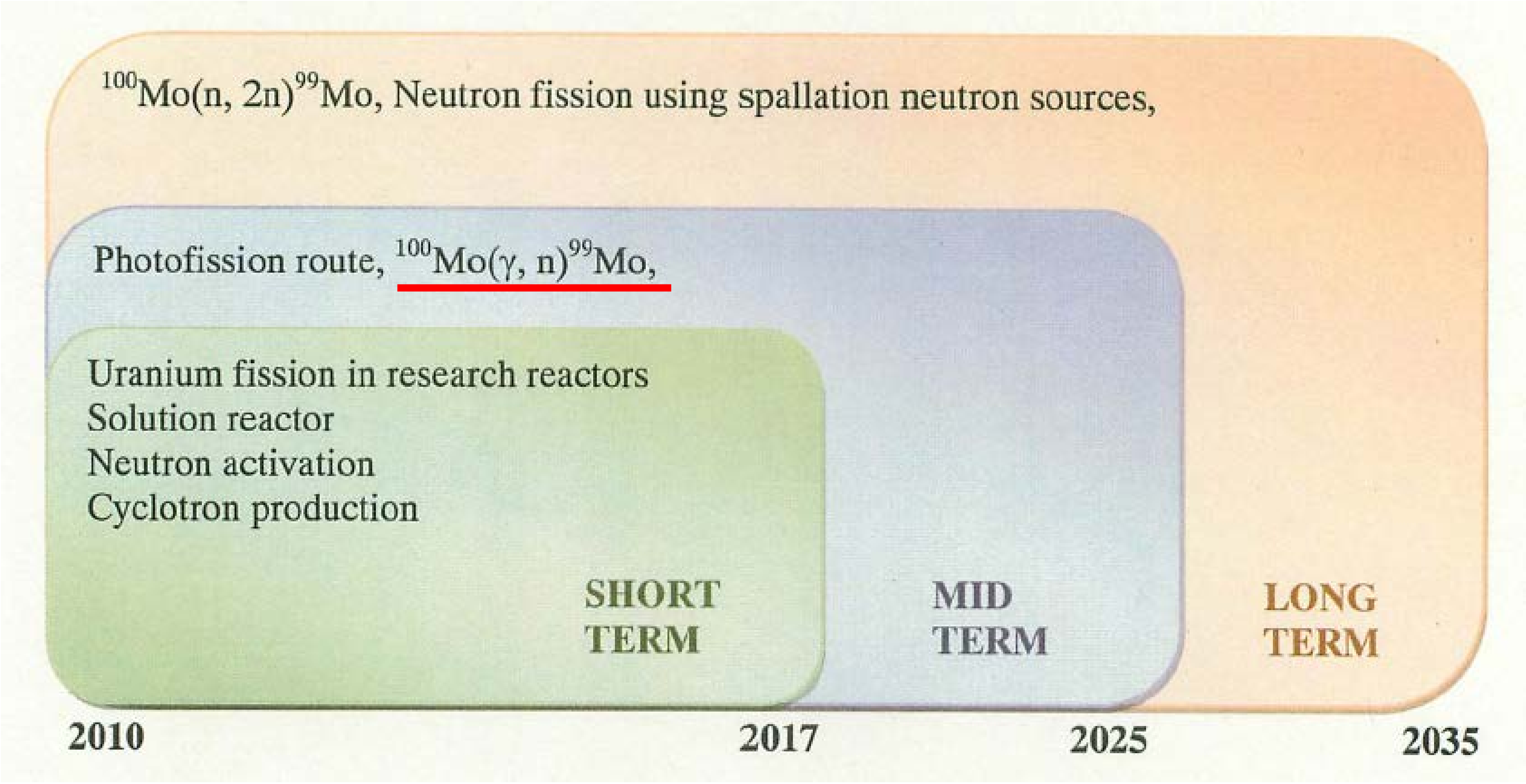


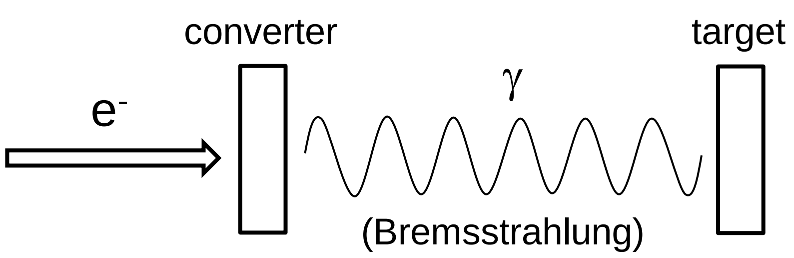
〇100-300 μA, 6 h照射で7000-21000 mCi　（300-900回の検査；25 mCi/1回）

〇ターゲット量：~ 0.2 g（直径6 mm，厚さ1 mm）

OECD/NEA report, 2010

制動放射線を用いた^{99}Mo製造

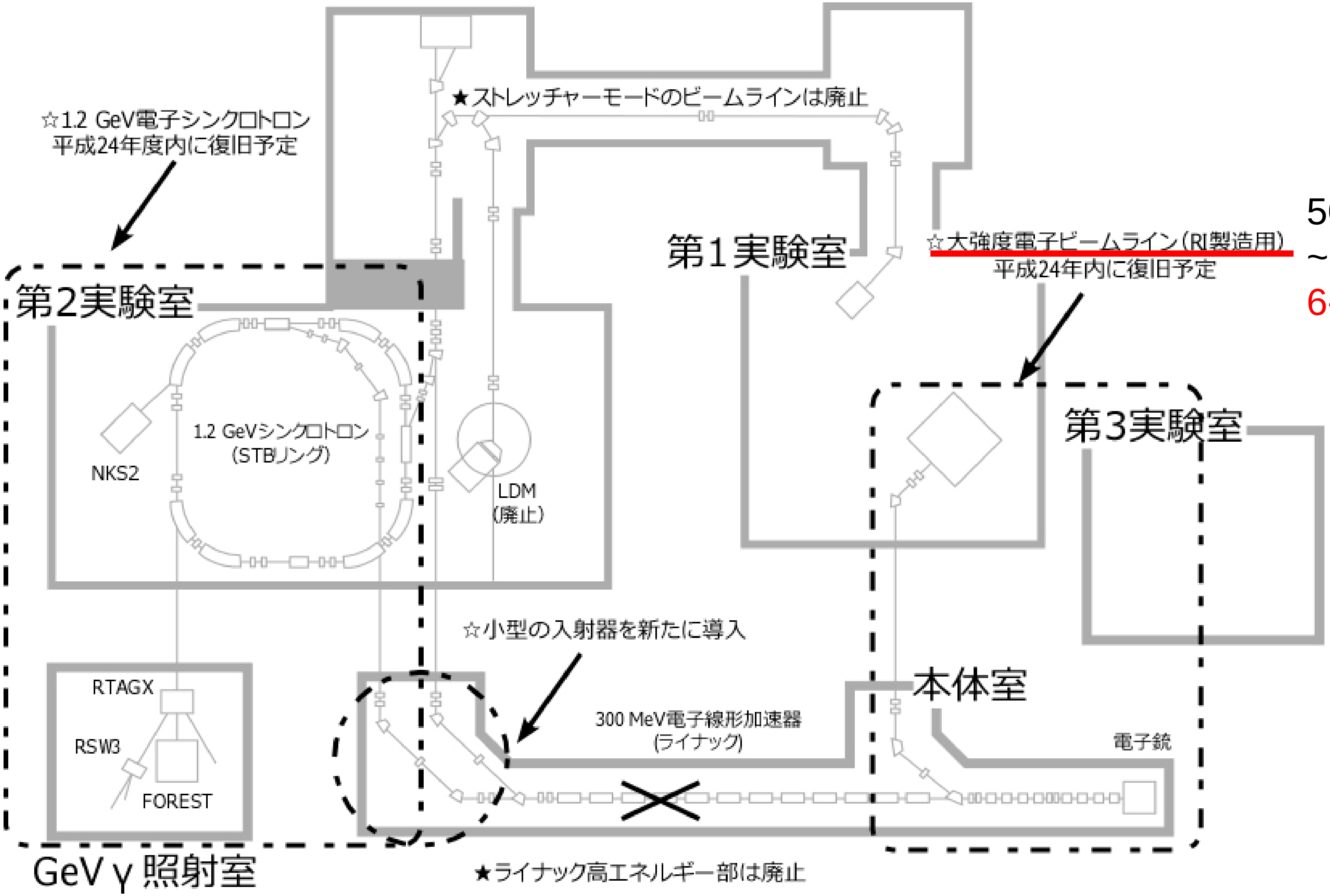




○ELPH(LNS)-Linac

○KUR-Linac

電子光ライナックの現状



50MeV
~120 μA
6-7 kW

制動放射線を用いた^{99}Mo製造

7.2 Reaction ^{100}Mo(γ,n)^{99}Mo

The cross section of ^{100}Mo(γ,n)^{99}Mo reaction is approximately equal to 0.1 barn for the energy interval 12-17 MeV. Thus, one can imagine a 25-50 MeV electron accelerator generating gamma rays via bremsstrahlung in a converter (same as for the photofission route described in Section 7.1), and using them for the (γ,n) reaction on a ^{100}Mo target.

The advantage of the ^{100}Mo(γ,n)^{99}Mo reaction is that its macroscopic cross section is considerably larger than in the photofission route. Comparing the U_3O_8 targets envisaged for the photofission production with ARIEL and metallic molybdenum targets highly enriched in ^{100}Mo, this ratio[36] is more than 35.

The production yield and the specific activity of molybdenum strongly depend on the type and dimensions of the target and converter. According to the recent calculations and measurements (TRIUMF, 2008 and Bunatian *et al*., 2009), about 650 6-day Ci of ^{99}Mo could be produced after 7 days of irradiation with an accelerator like ARIEL (500 kW, see Section 7.1) and a target of 30 g of highly-enriched molybdenum. The specific activity of molybdenum in this case would be ≈20 Ci/g.

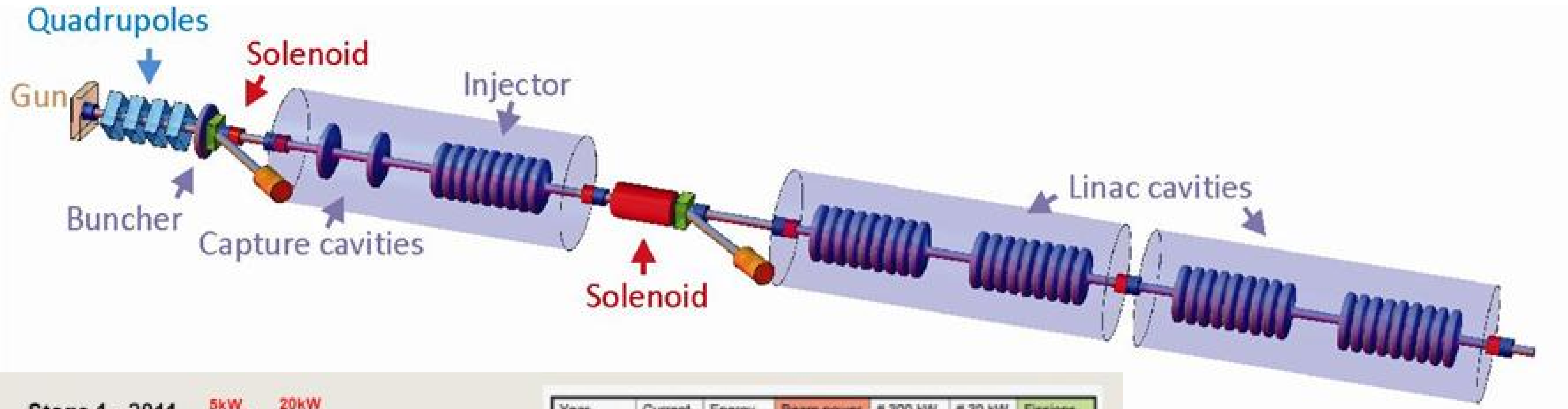
The ^{99}Mo production yield is higher in the ^{100}Mo(γ,n)^{99}Mo compared to the one in the photofission route. However, the maximum predicted specific activity would be of several hundred Ci of ^{99}Mo per gram of molybdenum (TRIUMF, 2008). This is low compared to the minimum specifications of about 1 000 Ci per g_{Mo} for existing commercial technetium generators[37]. Similar to the low- and medium-activity molybdenum from the neutron activation of ^{98}Mo targets, a system of distributed centres of centralised ^{99m}Tc production has been envisaged (Bennett *et al*., 1999).

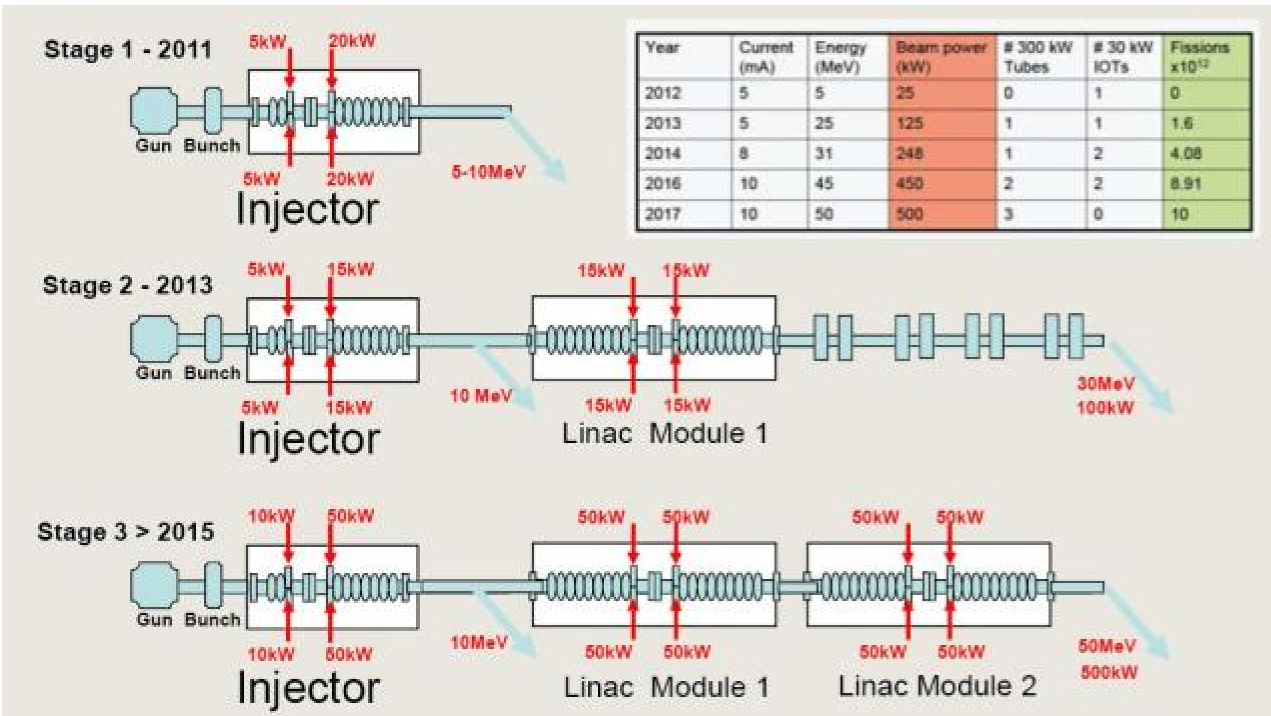
In Table 17, the ^{99}Mo production technology based on the ^{100}Mo(γ,n)^{99}Mo reaction is assessed according to the list of criteria defined in Section 5.3.

^{99}Mo：~40 Ci / week

OECD/NEA report, 2010

海外でのRI製造





Year	Current (mA)	Energy (MeV)	Beam power (kW)	# 300 kW Tubes	# 30 kW IOTs	Fissions $x10^{12}$
2012	5	5	25	0	1	0
2013	5	25	125	1	1	1.6
2014	8	31	248	1	2	4.08
2016	10	45	450	2	2	8.91
2017	10	50	500	3	0	10

2012年 : 25 kW

⇩

2017年 : 500 kW

TRIUMF/ARIEL HPより

問題点

○照射中の空間線量

中性子, γ線

○放射性同位元素使用数量

東北大・ELPH : ^{99}Mo,　40 MBq / 日
^{99}Tc^{m},　40 MBq / 日

東北大・CYRIC : ^{99}Mo,　400 MBq / 日
^{99}Tc^{m},　400 MBq / 日　　cf.)1 Ci = 37000 MBq

○^{100}Mo濃縮同位体

高価　⇨　再利用, 収率のよい化学分離法

マクロ量のターゲット, 副反応生成物

まとめ

○プロトン照射，制動放射線照射による$^{99}Mo/^{99}Tc^{m}$製造に関して

○現有施設でも照射装置，化学分離装置の改良で，有事の際に$^{99}Mo/^{99}Tc^{m}$供給が可能？

○最大使用数量が大きな問題
（ビーム量増強しても製造可能量が制限）