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Status of ^{99}Mo/^{99m}Tc Production Development by (n,γ) Reaction in JMTR

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1. Introduction

Background

Technetium-99m (^{99m}Tc) is one of commonly used radioisotopes in the field of nuclear medicine. In Japan, all of ^{99}Mo, a parent nuclide of ^{99m}Tc, are imported from foreign countries.

JAEA has a plan to produce ^{99}Mo by (n, γ) method in the JMTR, because of safety, nuclear proliferation resistance and waste management. However, the specific activity of (n, γ)^{99}Mo is very low, compared with that of (n, *f*)^{99}Mo.

The R&D on domestic ^{99}Mo/^{99m}Tc production was adopted as new project under Tsukuba International Strategic Zone last year.

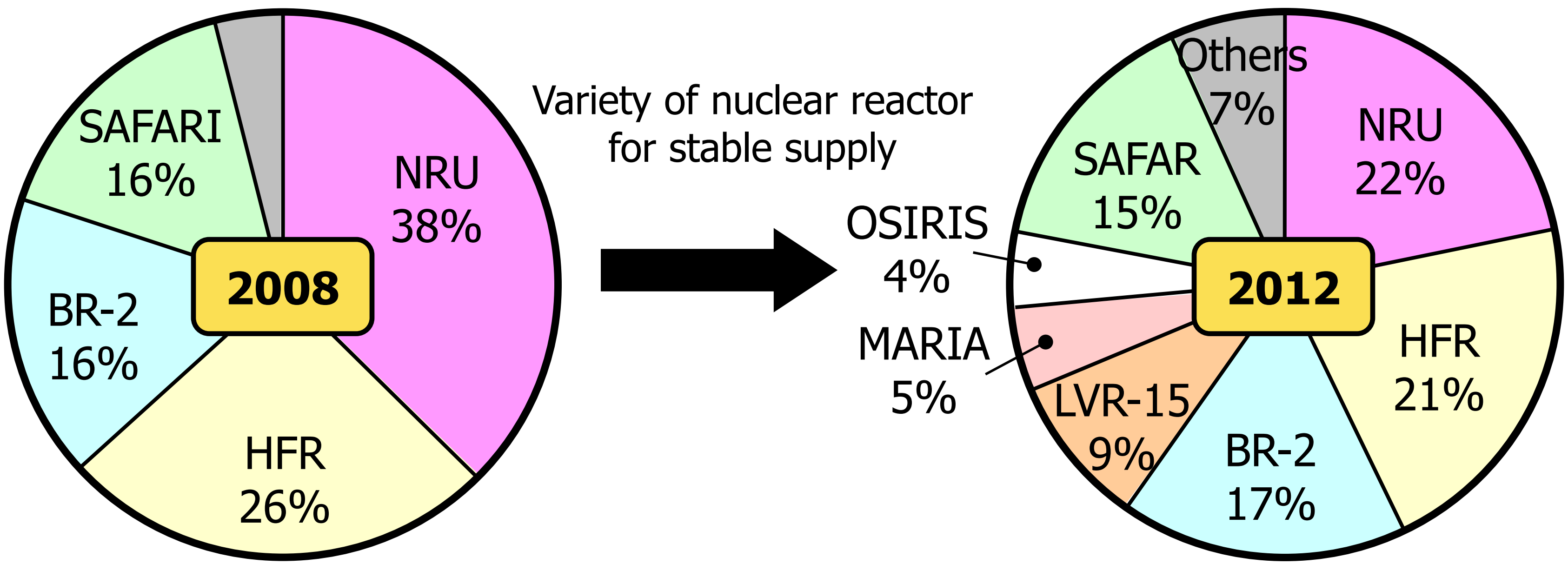
In this presentation, the R&D items and construction of the PIE devices are introduced under this project.

Issues for ^{99}Mo supply (1)

All of ^{99}Mo are imported from foreign countries in Japan.

- Stop of nuclear reactors for RI production due to accidents
- Air transport issues due to volcanic eruptions and other reasons

^{99}Mo production rate in overseas nuclear reactors



Ref. : NUCLEAR ENGINEERING INTERNATIONAL, JULY 2008

Ref. : OECD-NEA, A supply and Demand Update of the Molybdenum-99 Market, Aug. 2012

Status of nuclear reactor for RI production

1 : NRU in Canada will be shutdown at 2016.

2 : Difficult to construct the new reactors in Argentina, South Korea, etc. up to 2016.

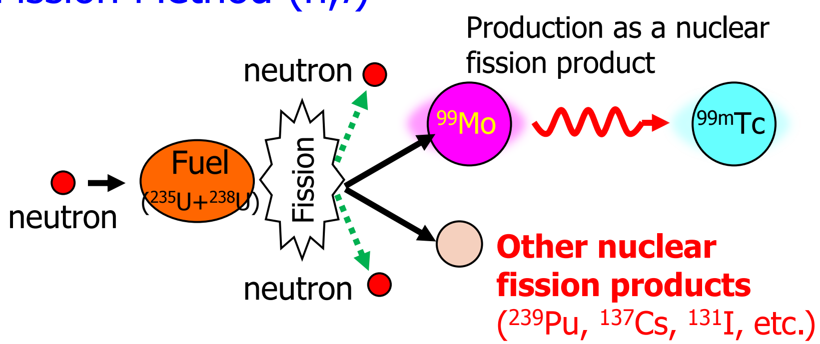
Issues for ^{99}Mo supply (2)

^{99}Mo is almost produced with enriched uranium.

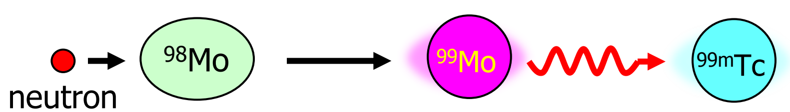
- Limited availability of high-enriched Uranium,
- Necessity of a technology similar to nuclear fuel recycling
- Necessity of management in term of non-proliferation and nuclear security

Production Methods of ^{99}Mo

Fission Method (n,*f*)



Neutron Activation Method (n,γ)



Comparison between Production Methods

	(n,*f*)	(n, γ)
Use of Uranium	**Yes**	**No**
Generation of Pu	**Yes**	**No**
Nuclear Proliferation Resistance	**Low**	**High**
Specific Activity	**High**	**(Low)**
^{99}Mo Production Cost	**High**	**(Low)**

Mar. 2012 : Seoul Nuclear Security Summit (Key actions)

1) Commitments from the governments of the four countries to support conversion of European production industries to non-HEU-based processes by 2015.
2) Support for U.S. exports of HEU to the European isotope producers to enable continued Mo-99 production until the facilities can convert to LEU targets.

Tsukuba International Strategic Zones

- A new industry-government-academia collaboration systems is to be constructed.
- The aim is to attain tangible results in five years from the pioneering projects in order to contribute to the growth and development of Japan in the fields of life and green innovation.

Seven Pioneering projects

Project 1 : Development and Implementation of Boron Neutron Capture Thrapy (BNCT)
Project 2 : Living with Personal Care Robots
Project 3 : Practical Use of Algal Biomass Energy
Project 4 : TIA-nano, Creating a global hub of nanotechnology
Project 5 : Development of Innovation Pharmaceuticals and Medical Technologies Using Biomedical Resources in Tsukuba
Project 6 : Domestic Production of Medical Radioisotope (Technetium-99m) in Japan
Project 7 : Achieving Practical Use of Revolutionary Robot Medical Equipment and Formation of a Global Focal Point

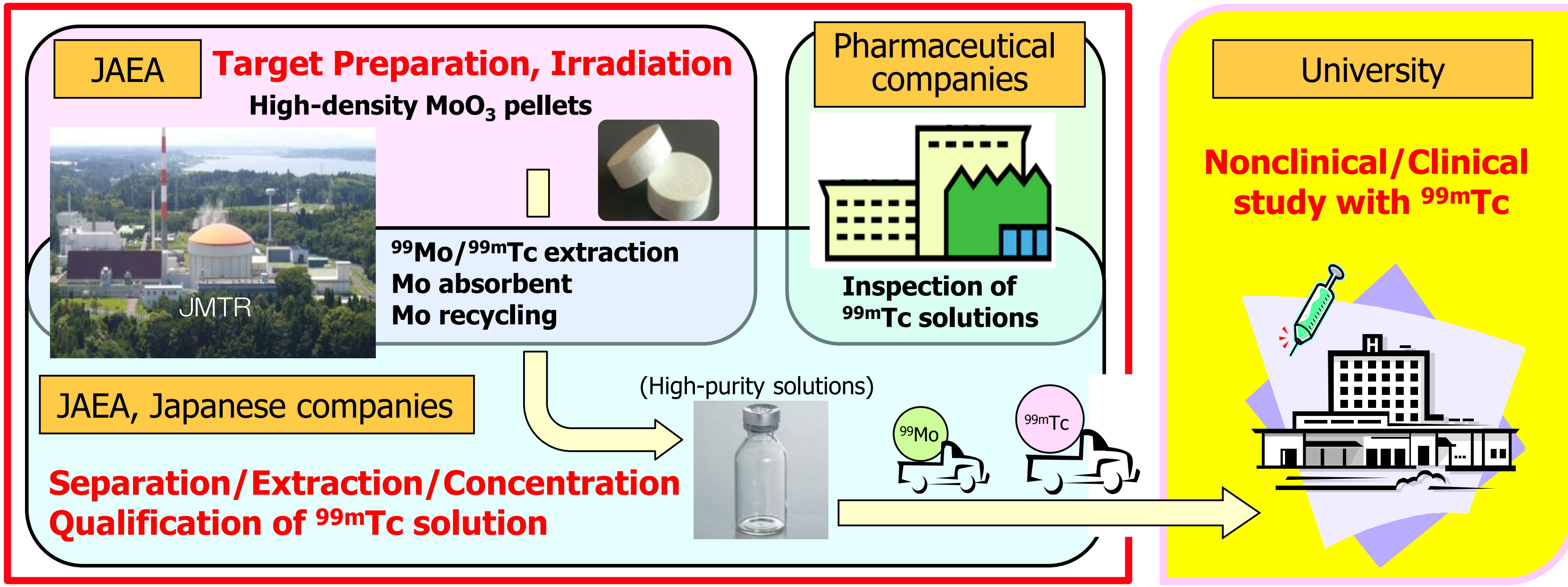
Our research and development (Project 6) has been received Initiatives in Tsukuba International Strategic Zones since October 2013.

R&D Items in Project 6

The following R&D in JMTR has been carried out for establishing the domestic $^{99}Mo/^{99m}Tc$ production by (n, γ) method.

Collaboration System in Project 6

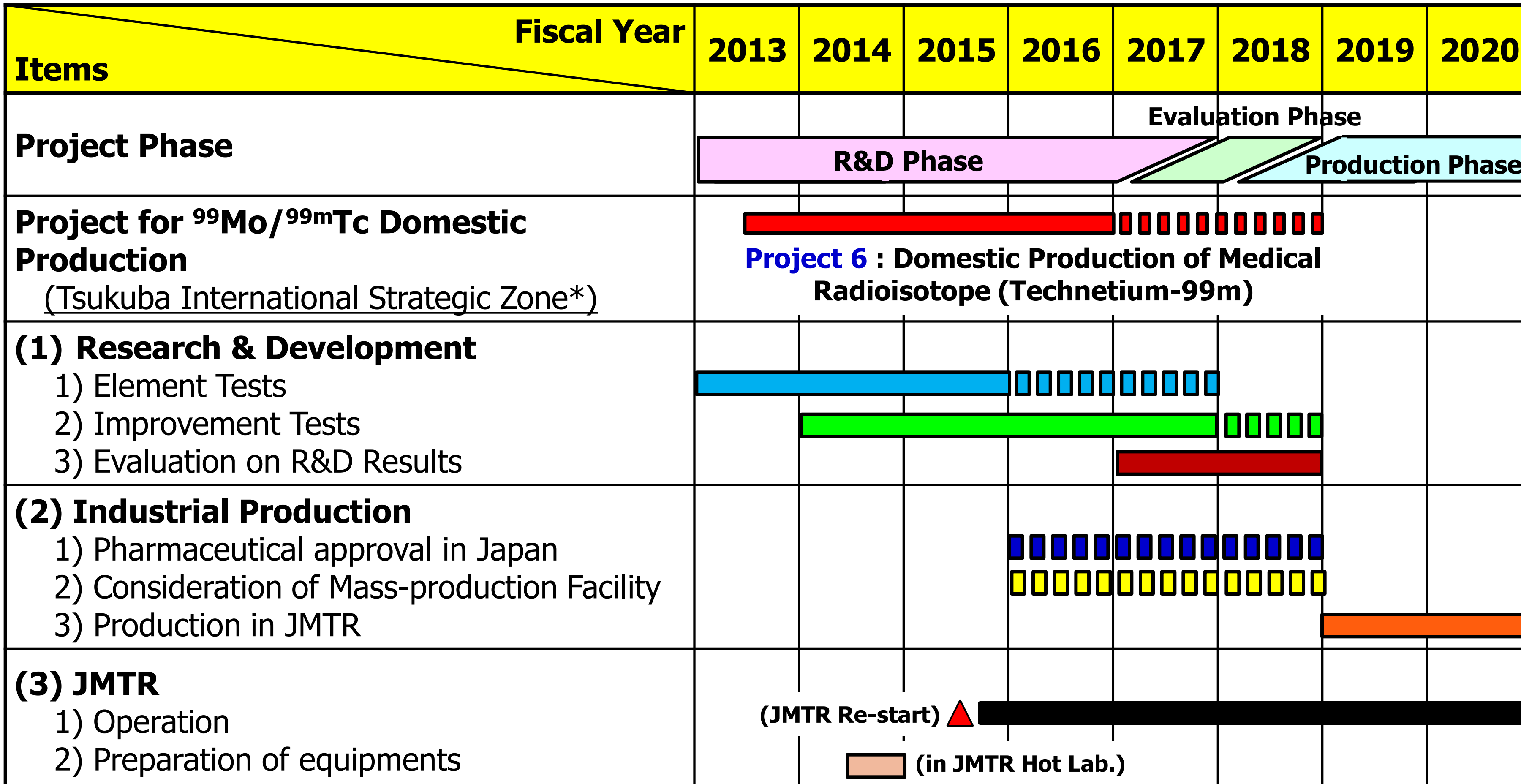
[Objects] Stable supply (Target : 20~25% in domestic demand)



(1) Fabrication of irradiation target such as sintered MoO_3 pellets
(2) Extraction and concentration of ^{99m}Tc by the solvent extraction from Mo solution
(3) Examination of ^{99m}Tc solution for medical use
(4) Mo recycling from the used Mo generators and waste Mo solutions

R&D Plan for Project 6

Each R&D item is carried out under the collaboration system of Tsukuba International Strategic Zone.



*** : The Japanese Prime Minister officially designated Tsukuba City and parts of Ibaraki Prefecture (Oarai-machi, etc.) as a Comprehensive Special Zone.**

2. R&D Items

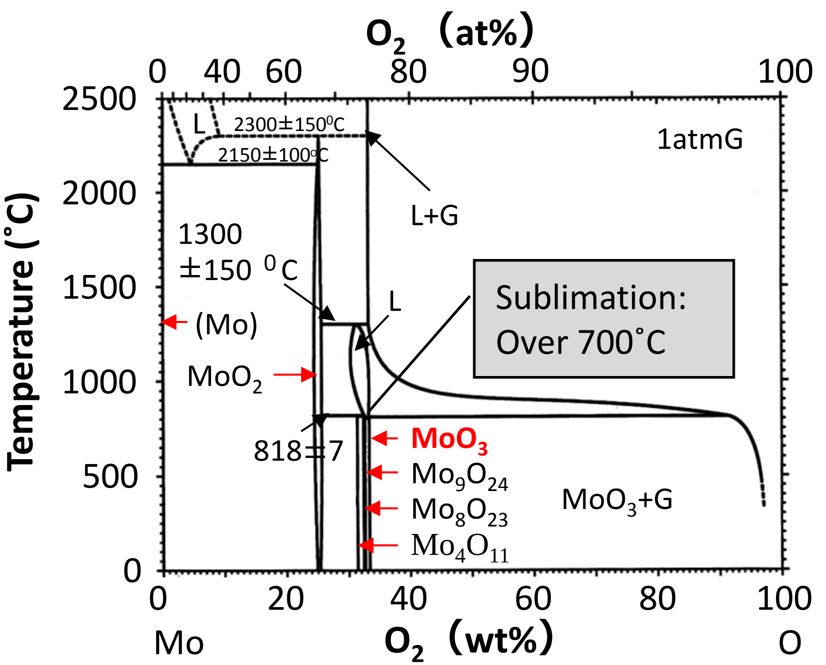
Fabrication Development for Irradiation Target

Metal Technology Co. Ltd.

It is necessary to fabricate the high density MoO_3 pellets (>90%T.D.) as an irradiation target for increasing of ^{99}Mo production for stable supply.

Properties of MoO_3

Density : 4.696 g/cm^3
Melting Point : 795˚C



Ref.: L.Brewer and R.H. Lamoreaux, 1980

Investigation of Fabrication Method

Cold press & Sintering (CS)
Plasma Sintering (PS)
Hot isostatic pressing (HIP)

Items \ Method	CS	PS	HIP
Mass Fabrication	F	P	E
Fabrication Time	F	F	E
Fabrication Cost	E	P	P
Density Control	P	E	E
Product Impurity	F	E	E

E : Excellent, **P** : Possible, **F** : Failure

It was successes to fabricate the high density MoO_3 pellets by **PS method**.
The **HIP method** is started for stable and mass fabrication of MoO_3 pellets.

Extraction and Concentration of Tc-99m

The (n, γ)^{99}Mo is **lower specific activity** than the fission ^{99}Mo. It is necessary for utilization by the (n, γ)^{99}Mo to develop the **extraction and concentration methods** of ^{99m}Tc solution through the extraction devices or ^{99}Mo/^{99m}Tc generators.

Irradiated MoO_3 Pellets

↓

Dissolution
(Mo Solution)

Master-Millker

(Solvent extraction)

^{99m}Tc

- **Separation/Extraction*** (with MEK)
- ↓
- **Concentration/Purification*** (Al_2O_3 column)
- ↓
- Conditioning (**^{99m}Tc solution**)

Column Method

(^{99}Mo/^{99m}Tc generator)

^{99}Mo

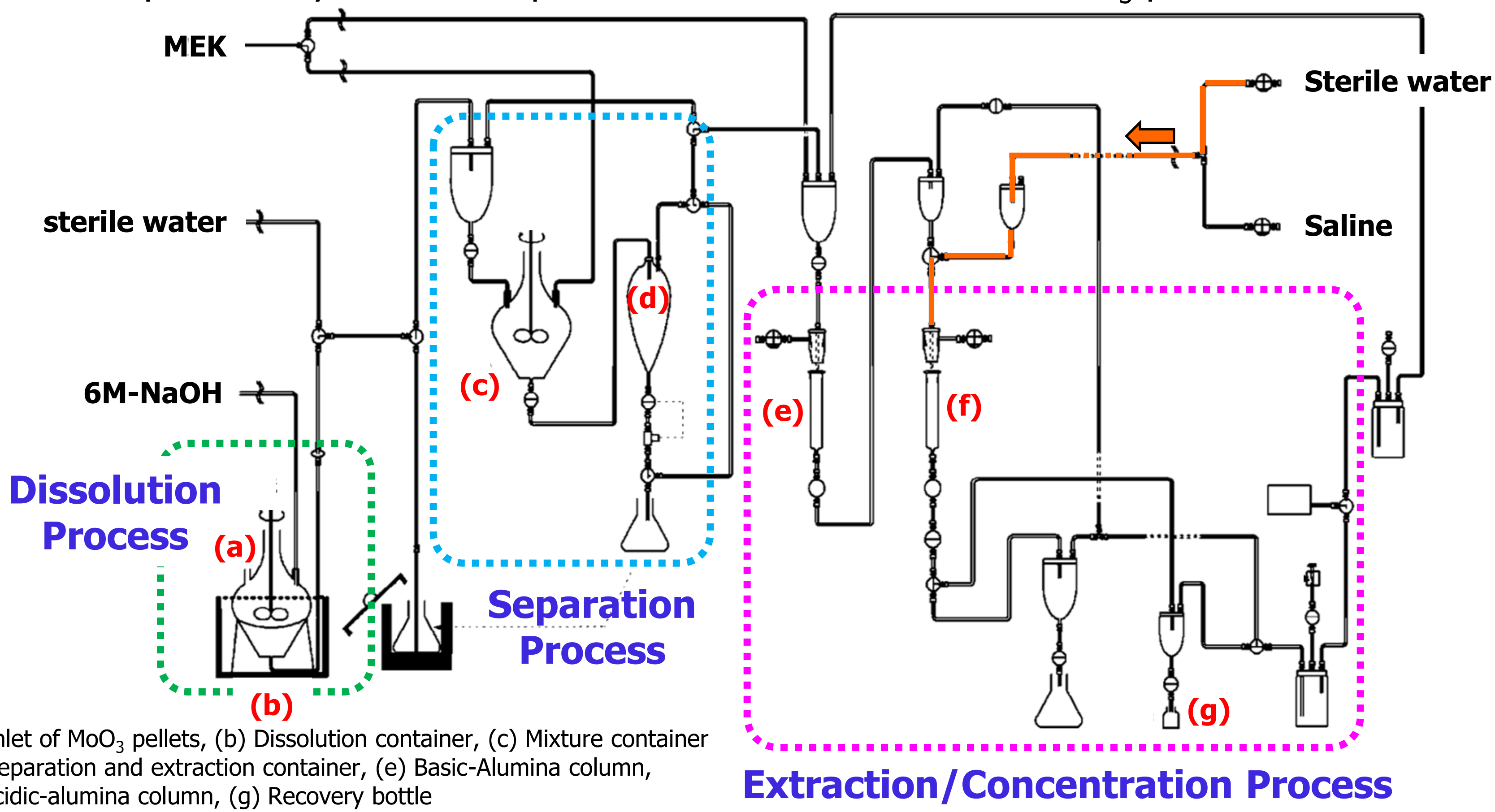
- **Separation/Extraction*** (Mo Adsorbent)
- ↓
- Washing/Assembly (Generator)
- ↓
- Conditioning (**^{99m}Tc solution**)

*** : R&D items**

R&D of Solvent Extraction for Master-Millker

The **solvent extraction** with MEK is still used for small local supply of ^{99m}Tc at several research reactor centers. Up to now, there has been no experience of a large scale ^{99m}Tc extraction by this method.

The $^{99}Mo/^{99m}Tc$ separation-concentration device was designed and constructed, and the preliminary tests were performed with the irradiated MoO_3 pellets in the JMTR.



(a) Inlet of MoO_3 pellets, (b) Dissolution container, (c) Mixture container
(d) Separation and extraction container, (e) Basic-Alumina column,
(f) Acidic-alumina column, (g) Recovery bottle

The **improved device** is designed for mass extraction of ^{99m}Tc solution.

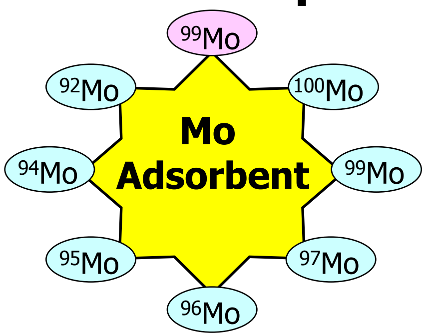
R&D of Mo Adsorbent for $^{99}Mo/^{99m}Tc$ Generators

ART KAGAKU Co., Ltd.

It is necessary for $^{99}Mo/^{99m}Tc$ generators loaded with (n, γ)^{99}Mo to develop the Mo adsorbents with high Mo absorption amounts. Some Mo adsorbents have been fabricated and the characterization of these adsorbents was carried out.

(n, γ)^{99}Mo

1) Lower specific activity
2) Stable isotopes in Mo after irradiation



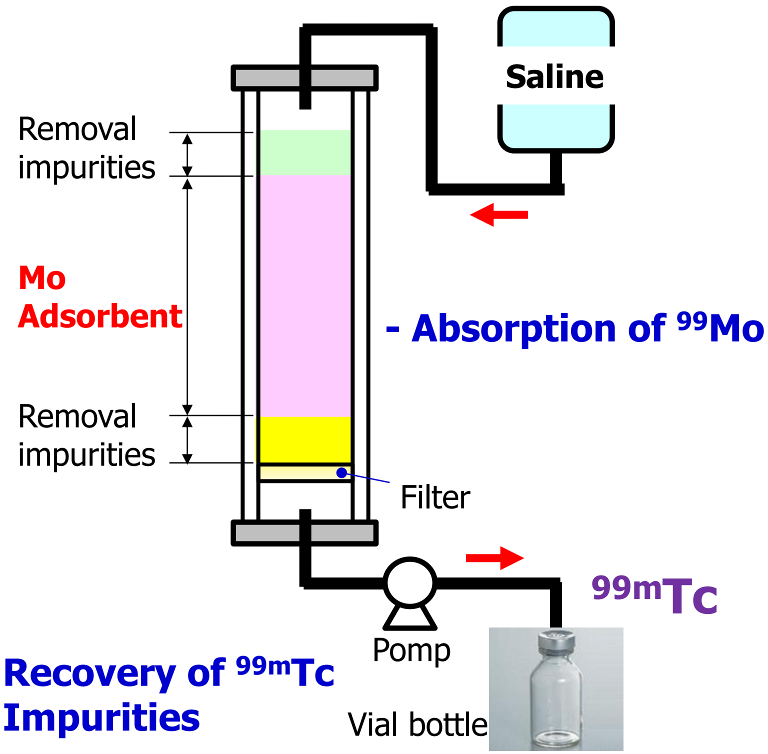
High Mo absorption amounts

Candidate Mo Adsorbents

Mo Adsorbent	Mo absorption amounts
Al_2O_3	2~20 mg/g
PZC	250 mg/g
PTC	250 mg/g

PZC : Polyzilconium compound, **PTC** : Polytitanium compound

Experimental for Column Design



- Recovery of ^{99m}Tc
- Impurities

The **improved Mo adsorbent** is developed and characterization is carried out for production of high-purity ^{99m}Tc solution.

Development of Mo Recycling

Taiyo Koko Corporation

MoO_3 is used as the irradiation target for the production by (n, γ)^{99}Mo. Recycling technology development is proposed to recover molybdenum (Mo) for the effective use of resources and reduction of radioactive wastes.

Subjects for Mo Recycling

(a) Recovery of Mo resource

Mo recovery from the Mo solution and used ^{99}Mo/^{99m}Tc generators (PZC, etc.) and utilization of recovered Mo.

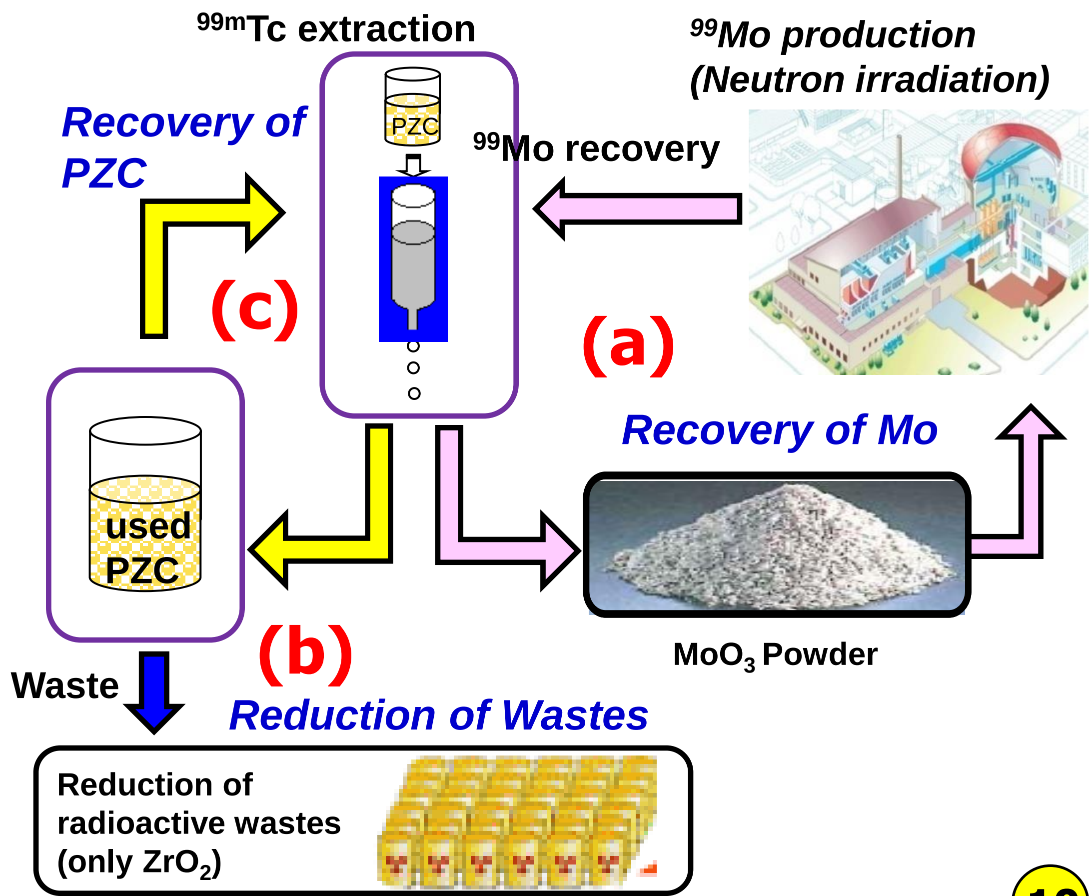
(b) Reduction of Wastes

Treatment and waste reduction of used ^{99}Mo/^{99m}Tc generators (PZC, etc.) and Mo solution

(c) Recovery of Mo adsorbent

Development of reusable ^{99}Mo/^{99m}Tc generators

Recycling System with PZC (for example)



Specification of ^{99m}Tc solution for Medicine

The specifications of ^{99m}Tc solution are determined in pharmacopoeia in each country. It is important for domestic production to determine detail specifications of ^{99m}Tc solution.

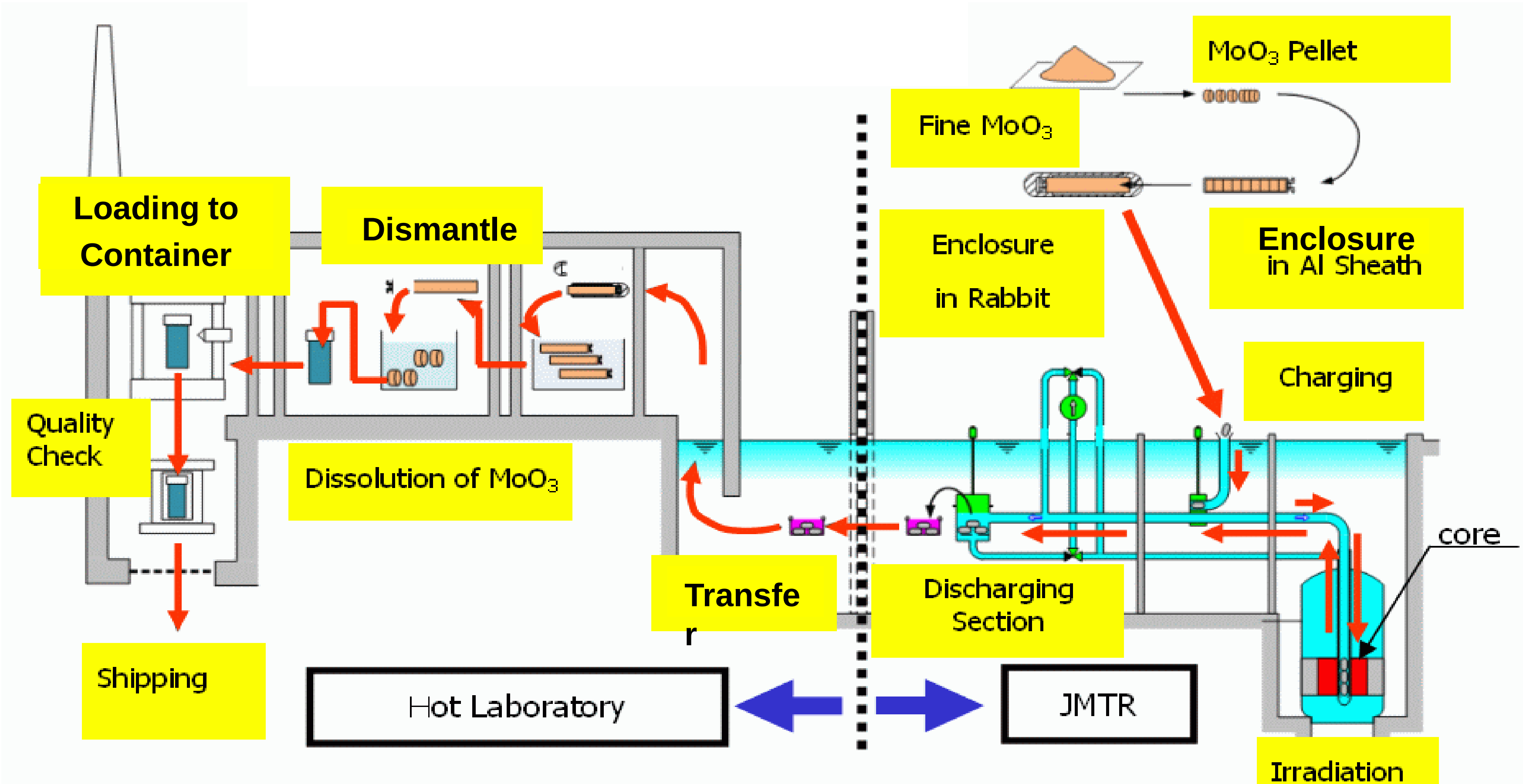
Items	JPN Pharmacopeia	(*n, fiss*) by USP	(*n, γ*) by USP
^{99m}Tc content (%)	90 ~ 110	90 ~110	90 ~110
Radiochemical purity (%)	≥ 95	≥ 95	≥ 95
Radionuclidic impurity (μCi/mCi ^{99m}Tc)			
^{99}Mo	≤ 0.15	≤ 0.15	≤ 0.15
^{131}I	—	≤ 0.05	—
^{103}Ru	—	≤ 0.05	—
^{89}Sr	—	≤ 0.006	—
^{90}Sr	—	≤ 0.00006	—
Others	—	β&γ emitters≤0.1 α Emitter≤$1x10^{-6}$	≤ $1x10^{-5}$
pH	4.5 ~ 7.0	4.5 ~ 7.5	
Chemical impurity			
Al	≤ 10 ppm	≤ 10 μg/ml	≤ 10 μg/ml
MEK	—	—	≤ 0.1%

USP : United States Pharmacopeia

3. Constructions of Irradiation and PIE Facilities

Flow Chart of $^{99}Mo/^{99m}Tc$ Production in JMTR

The ^{99}Mo production process has been developing in JAEA. The performance tests are also carried out in each part. Now, the irradiation and PIE facilities are prepared.



PIE Facility
Environment Equipments of hot cells

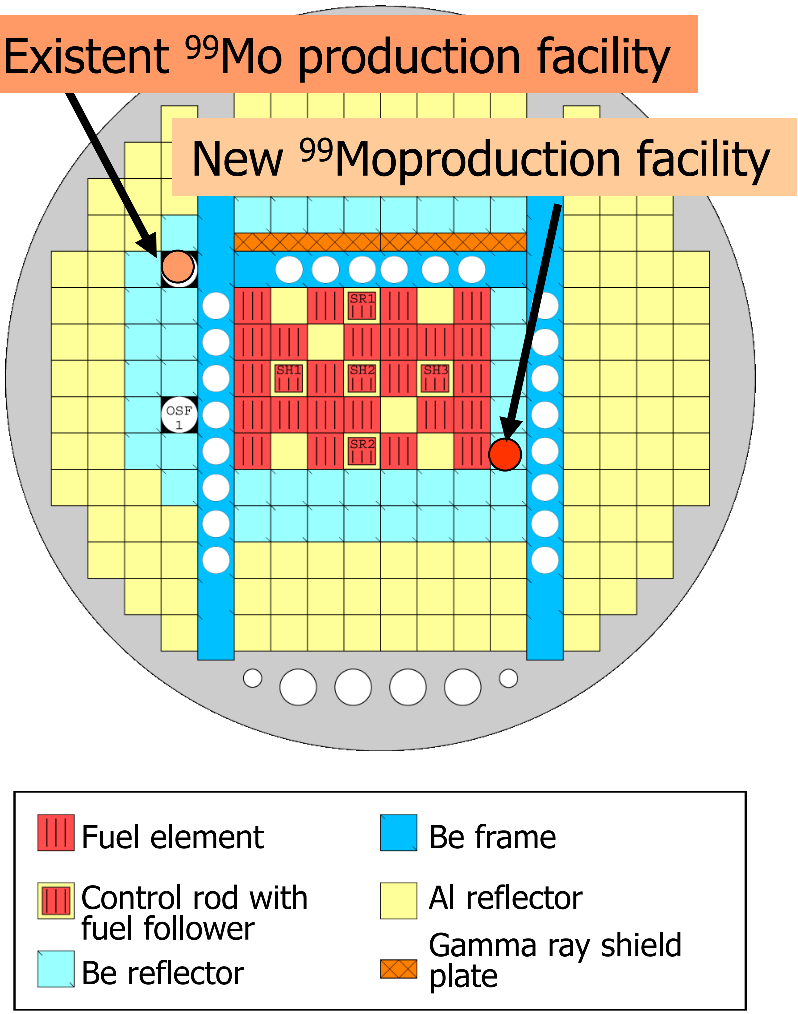
Irradiation Facility
Hydraulic rabbit Irradiation Facility

JMTR Core and ^{99}Mo Production Facility

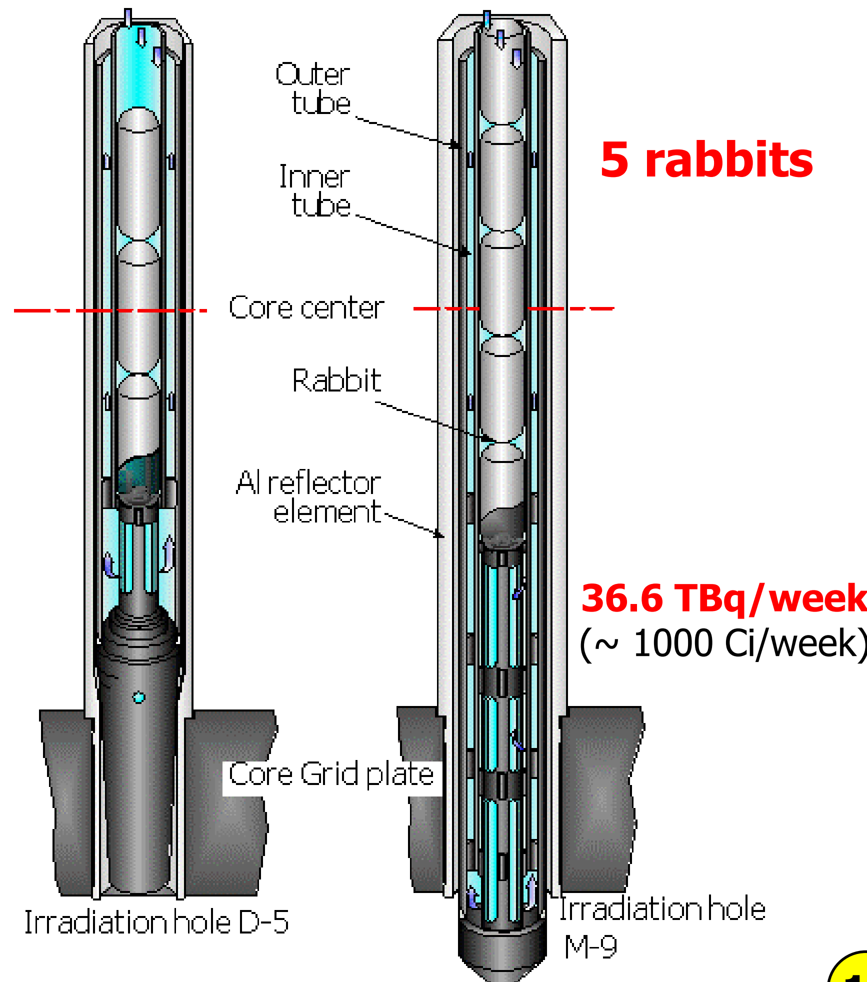
JAEA

The hydraulic rabbit irradiation facility will be used for the demonstration tests of ^{99}Mo/^{99m}Tc production in JMTR. The structure of new hydraulic rabbit will be revised.

Irradiation Holes



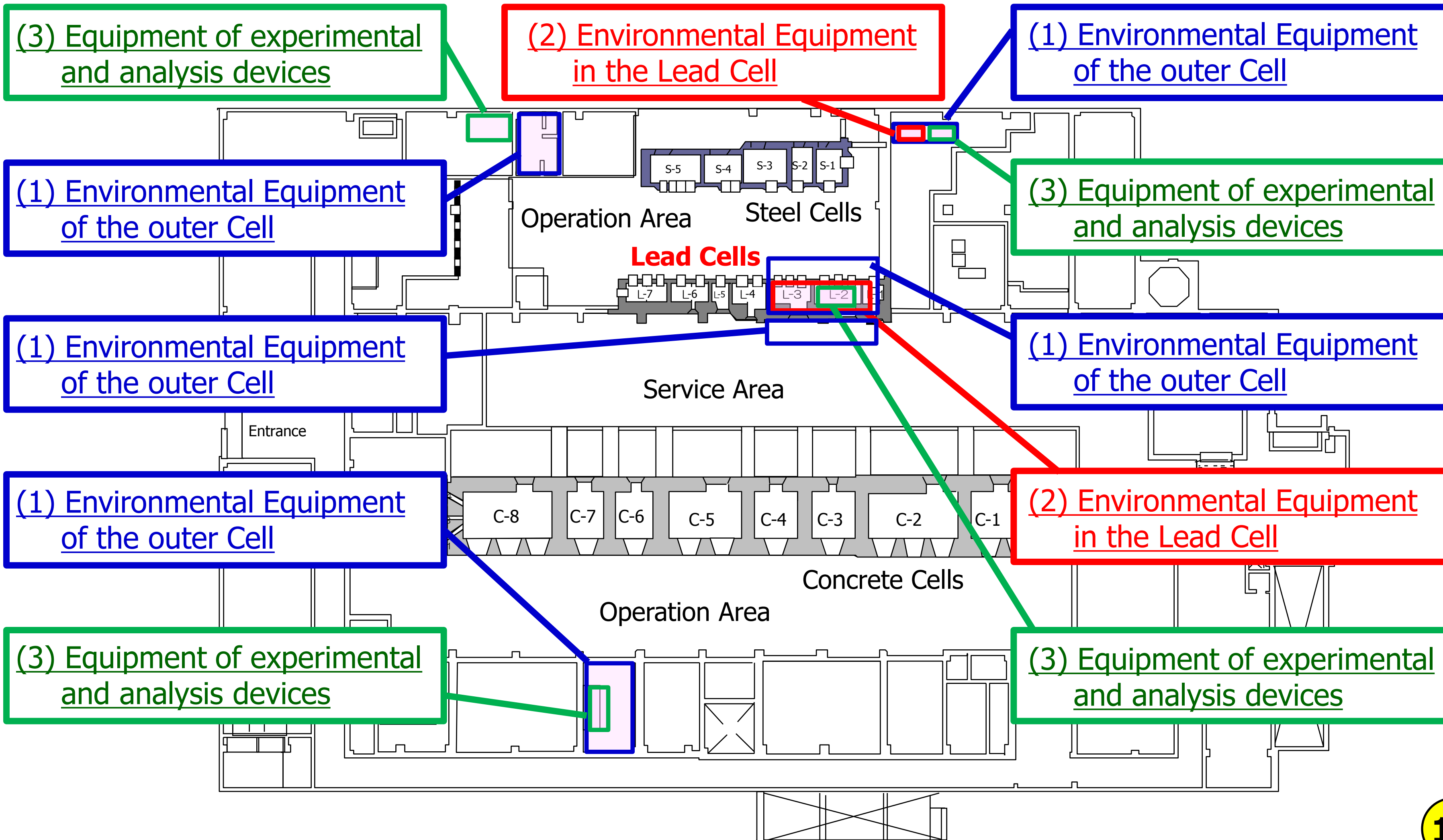
Structure of hydraulic rabbit tube



PIE Facilities for ^{99}Mo/^{99m}Tc Production

JAEA

The PIE facilities in the JMTR/HL will be equipped for the demonstration tests of ^{99}Mo/^{99m}Tc production in JMTR. The equipments are performed for environmental conditions.



4. Conclusion

Conclusions

^{99}Mo production by (n, γ) method has been determined from viewpoints of nuclear nonproliferation, reduction of radioactive wastes and utilization of Mo resource in JMTR.

- The sintered MoO_3 pellet with high density will be used as irradiated target material for ^{99}Mo production and fabrication development of high density MoO_3 pellet is carried out.

- Development of $^{99}Mo/^{99m}Tc$ extraction and concentration methods is carried out and useful data are accumulated under the collaboration system of Tsukuba International Strategic Zone.

- The extracted ^{99m}Tc solution is examined according to the pharmacopoeia and nonclinical/clinical study with ^{99m}Tc solution will be performed.

- The facilities in the JMTR and JMTR/HL are prepared for the demonstration tests of $^{99}Mo/^{99m}Tc$ production.