

茨城大学大学院理工学研究科
「原子科学と倫理」
2011年12月27日(火)
3時限13:00-14:30

核燃料サイクルの資源戦略

Resourceability on Nuclear Fuel Cycle

Masaki Ozawa

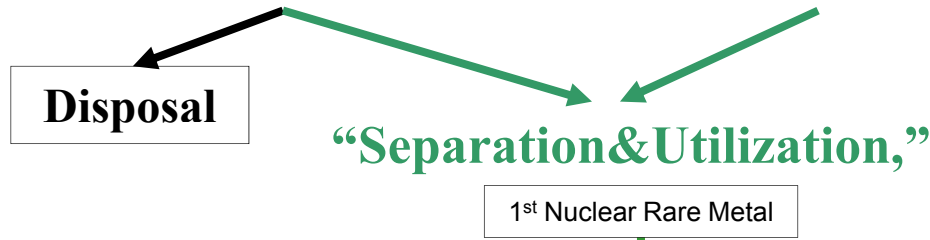
Tokyo Institute of Technology
JAPAN



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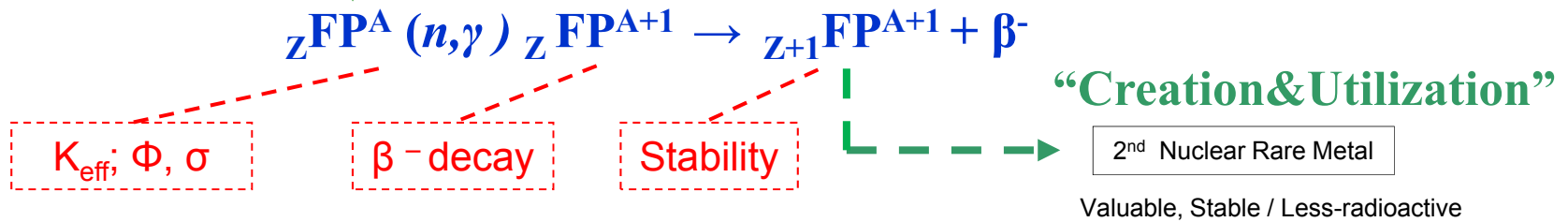
1/ Advanced ORIENT Cycle

^{235}U (n,vn) FP , Energy ($7.8 \times 10^7 \text{KJ}/^{235}\text{Ug}$)



- The 389th Session of Xiangshan Science Conference (香山科學會議); Radiochemistry of Nuclear Fuel Reprocessing
- 22-24, December 2011
- Beijing, China

2/ Après ORIENT



Nuclear Creation of Rare Metals

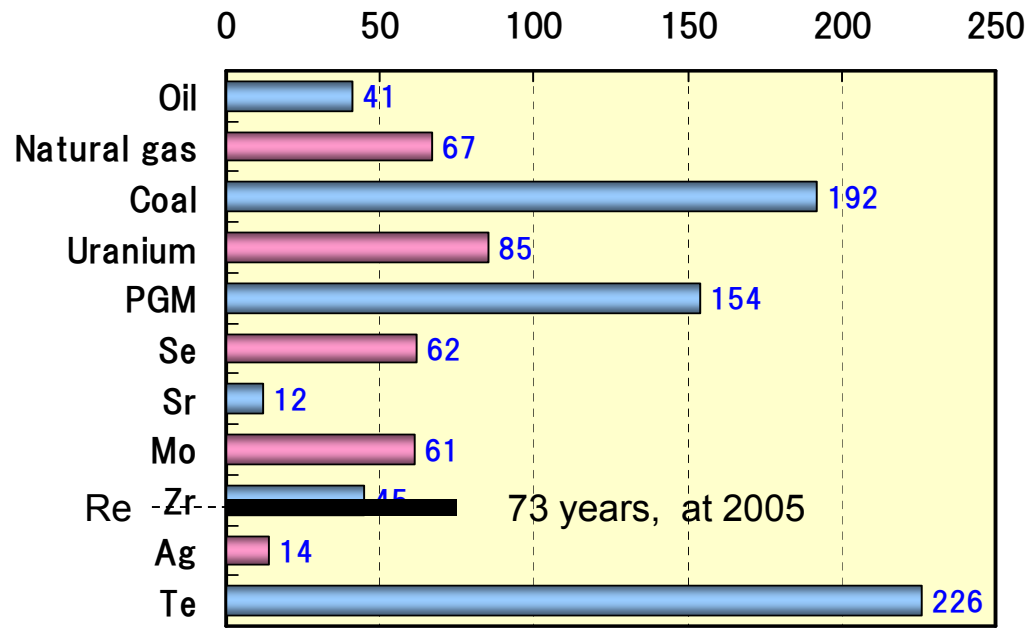
Definition as Rare Metals in Japan ; 47 elements, including 17 rare earth

Definition as Nuclear Rare Metals (tentative) ;31 elements in $>10\text{g/t}$,

e.g.,excluding Noble gas, Halogen, Cd, Sn, Sb, Bk, Cf

Group	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	
Period 1	1 H			FP														2 He	
Period 2	3 Li	4 Be												5 B	6 C	7 N	8 O	9 F	10 Ne
Period 3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar	
Period 4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr	
Period 5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe	
Period 6	55 Cs	56 Ba	57 La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn	
Period 7	87 Fr	88 Ra	89-103 Ac	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt										
Lanthanides	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu				
Actinides	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr				

Incentives; Energy and Rare Metals(Earth) for National Security

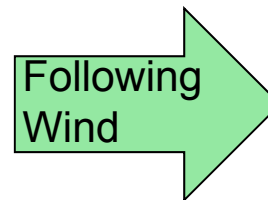


R/P Ratio (year) at 2004 on Estimated Available Time

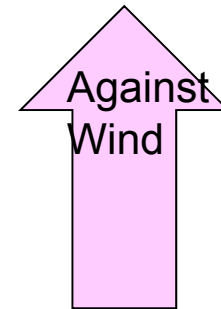
■ PGM (ca.200y) and Rare Earth (RE) (>850y) seems to be rather abundant however, they are extremely localized. Namely, ca.90% of RE were from China (Japanese case), and about 1% of national GDP will drop, if ca.20% of those supply decreased.

南巡講話・鄧小平1992.1 ”中東有石油、中国有稀土”
Rare Metal (Earth) strategy will dominate the national security at every countries, as the same as Energy.

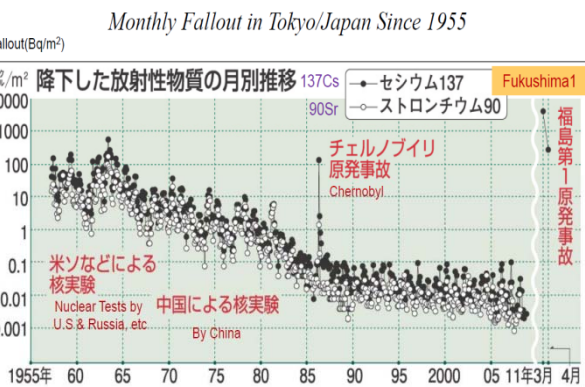
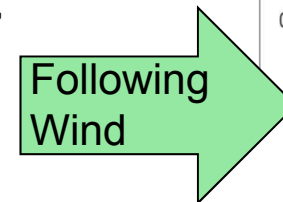
- Natural fossil fuels (Oil, Gas) and U are limited to 40-80 years, and Coal is limited to around 200 years.
- Worldwide CO₂ issue (waste of Oil !) is inevitable.



Nuclear (FBR) Renaissance in Europe, USA, (Japan), etc



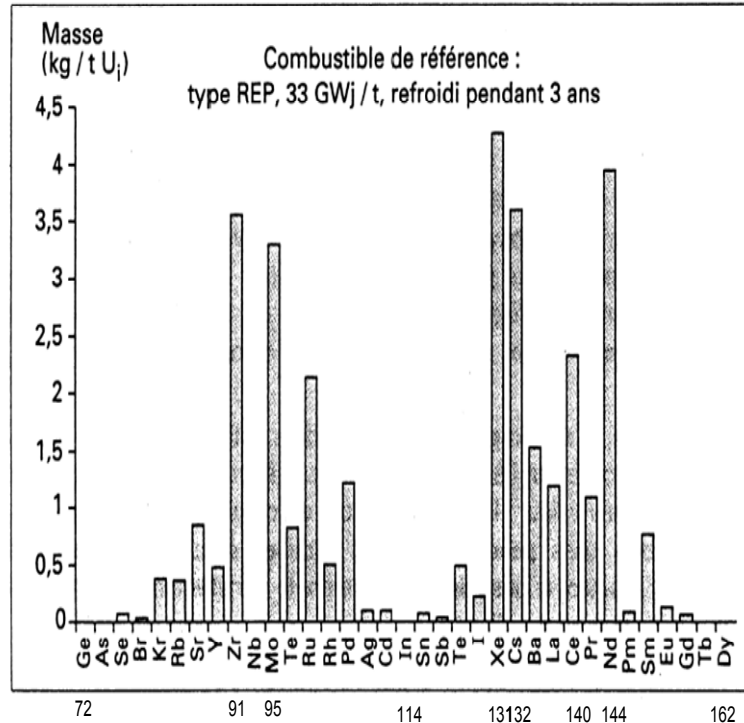
3.11 Catastrophe of Fukushima NPP in Japan



Urban Mine, Nuclear Ore

放射性廃棄物から白金族 (Ru,Rh,Pd) 、レアアースの回収 (原子力鉱山)

FBR MOX, Inner core, 150GWd/t, cooled 5 years



高燃焼FBR

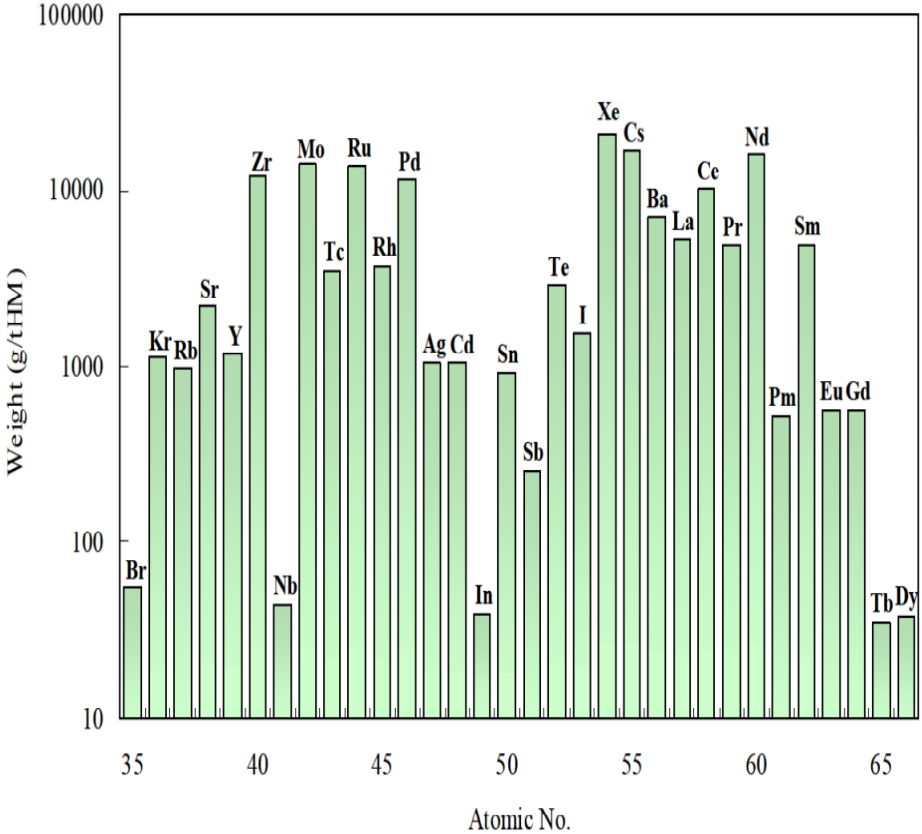


Figure - Masse des éléments chimiques constituant les produits de fission

Mass of FP elements in irradiated fuel

(M.Bourgeois, Retraitement du combustible, 1994)

軽水炉燃料再処理工場800 t /y, 回収率80 %とすると

原子力レアメタルの生産量; Tc: 0.64t, PGM: 2.56t, Lns: 6.4t, Mo:1.92t, 他

Value of Synthesized HLLW ca.1975\$(148,100¥) / L (HCl-HLLW)

ca.5700\$(426, 520¥) / L (HNO₃-HLLW)

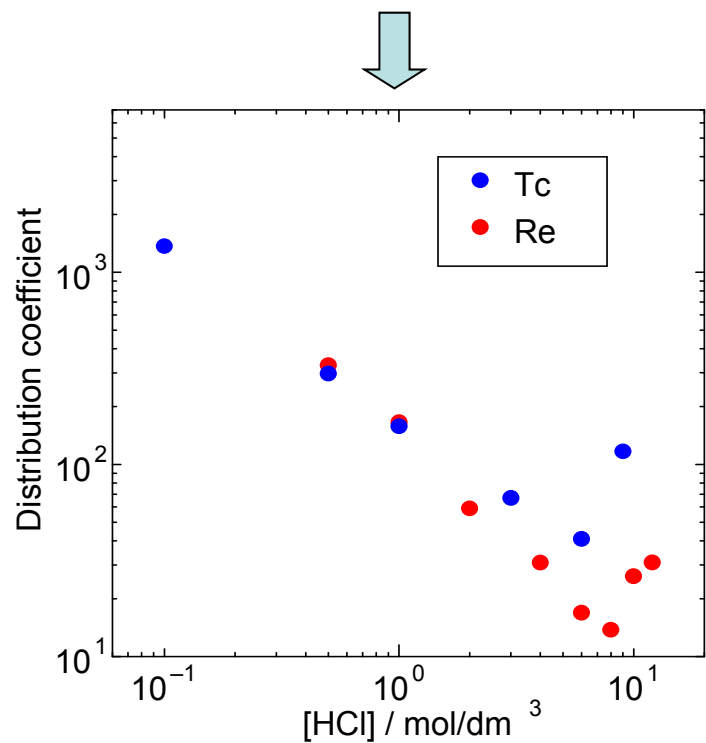
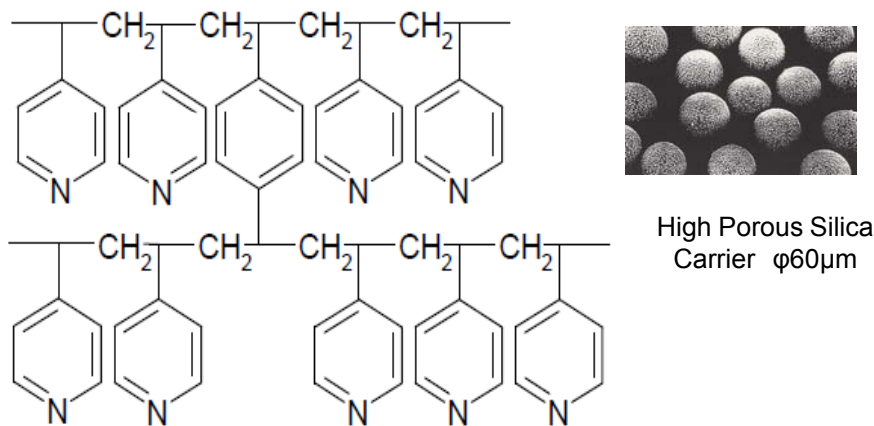
No.	Element	Conc. (g/L)	Chemical Formula	Grade	¥/g	\$/g
1	Fe	4.340	FeCl ₂ ·4H ₂ O	特級	6.6	
2	Cr	0.205	CrCl ₃ ·6H ₂ O	鹿特級 >93.0%	29	
3	Ni	0.550	NiCl ₂ ·6H ₂ O	特級 >98%	68	
4	Rb	0.311	RbCl	>95%	940	
5	Cs	2.140	CsCl	>98%	132	
6	Sr	0.769	SrCl ₂ ·6H ₂ O	特級 >99%	68	
7	Ba	1.340	BaCl ₂ ·2H ₂ O	特級 >99%	56	0.7
8	Zr	3.290	ZrCl ₄	>99.5%	820	
9	Mo	2.940	MoCl ₃	99.5%	4,950	66
10	Re	0.876	HReO ₄	76.5%水溶液	3,400	45
11	Ru	1.710	RuCl ₃ ·3H ₂ O	>99.9%	3,500	47
12	Rh	0.349	RhCl ₃ ·3H ₂ O	-	35,000	467
13	Pd	0.921	PdCl ₂	特級	3,120	42
14	Ag	0.037	AgCl	99.50%	980	13
15	Cd	0.052	CdCl ₂ ·2.5H ₂ O	鹿特級	76	1
16	Sn	0.039	SnCl ₂ ·2H ₂ O	特級 >97.0%	56	
17	Se	0.043	Se ₂ Cl ₂	—	2,040	
18	Te	0.456	TeO ₂	>99.0%	140	
19	Y	0.433	YCl ₃ ·6H ₂ O	99.99%	260	3.5
20	La	1.100	LaCl ₃ ·7H ₂ O	>95%	72	1
21	Ce	2.110	CeCl ₃ ·7H ₂ O	>99%	116	1.5
22	Pr	1.050	PrCl ₃ ·7H ₂ O	>99.95%	340	5
23	Nd	3.610	NdCl ₃ ·6H ₂ O	>99.95%	380	5
24	Sm	0.767	SmCl ₃ ·6H ₂ O	>99.95%	280	3.7
25	Eu	0.121	EuCl ₃ ·6H ₂ O	>99.95%	2,200	29
26	Gd	0.061	GdCl ₃ ·6H ₂ O	99.9%	2,200	29
27	HCl	2M	HCl	特級35.0-37.0%	1.7	
Ref	Au				4,683	62
	Tc					ca.100

2nd Transition Elements (4d)

3rd Transition Elements(4f)

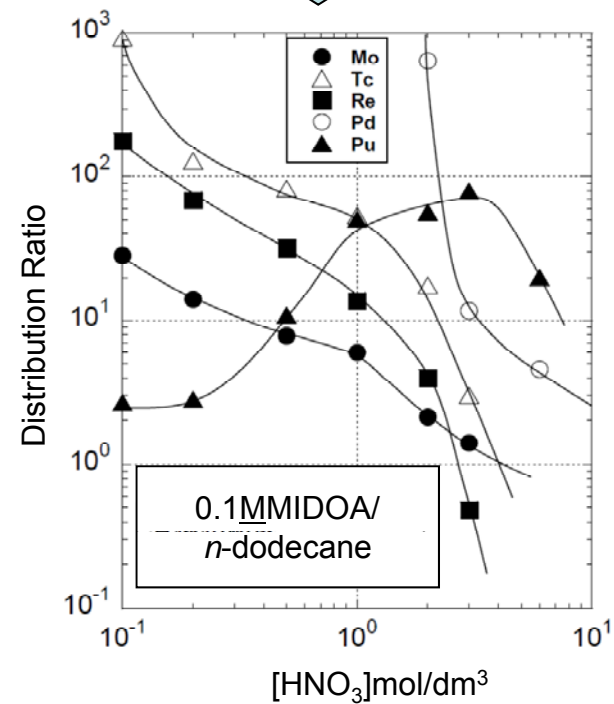
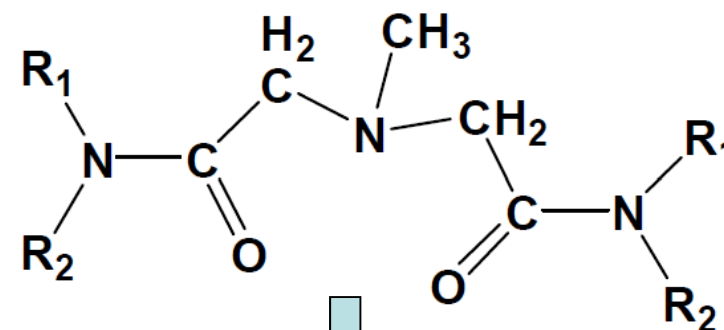
イオン交換クロマト分離法 (IXC) 及び 溶媒抽出法 (SX)

Tertiary Pyridine Resin (TPR)

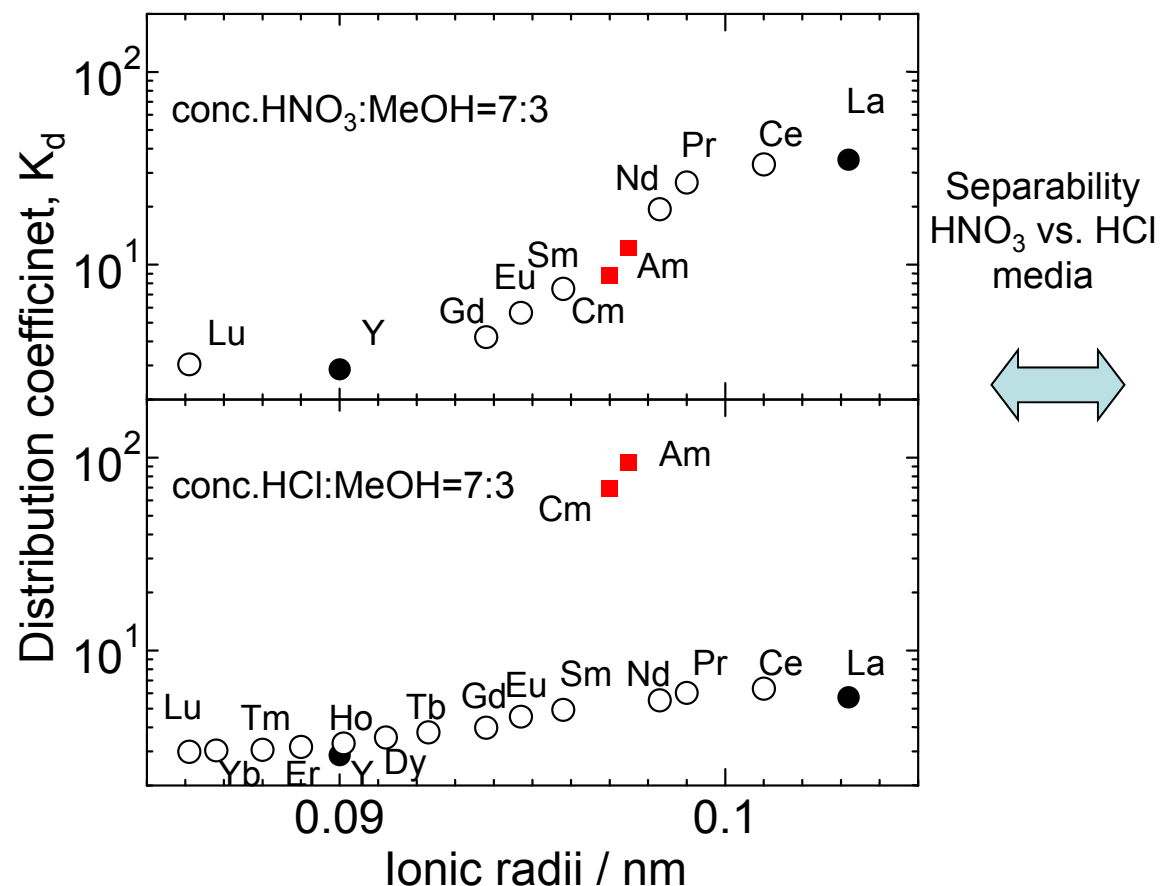


MIDOA

(Y. Sasaki, et.al., *Chem. Lett.* **36**, 1394 (2007).)



3級ピリジン樹脂のf元素の分配特性

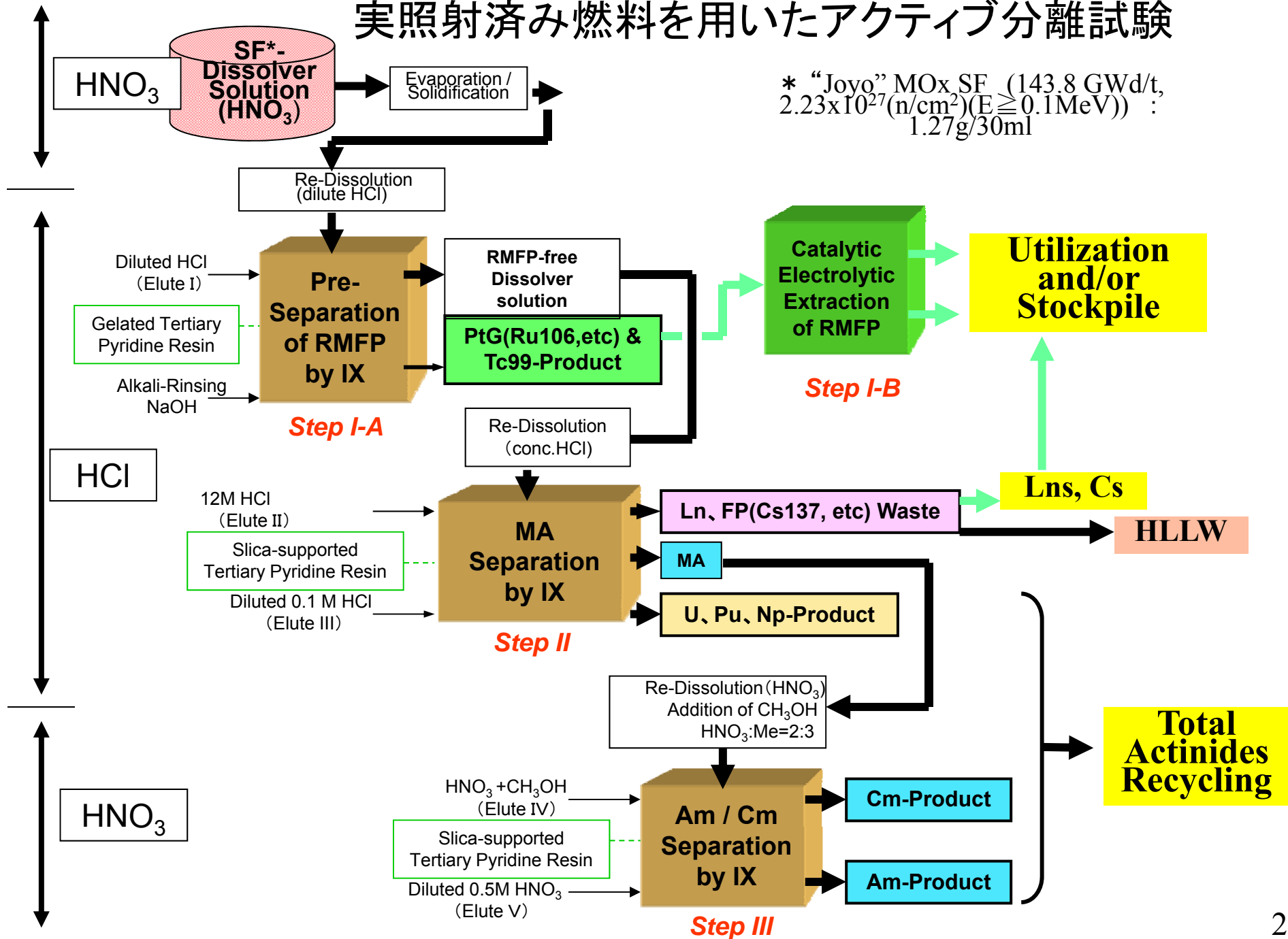


・HNO₃ 中では, 分離係数はイオン半径のみに依存する。

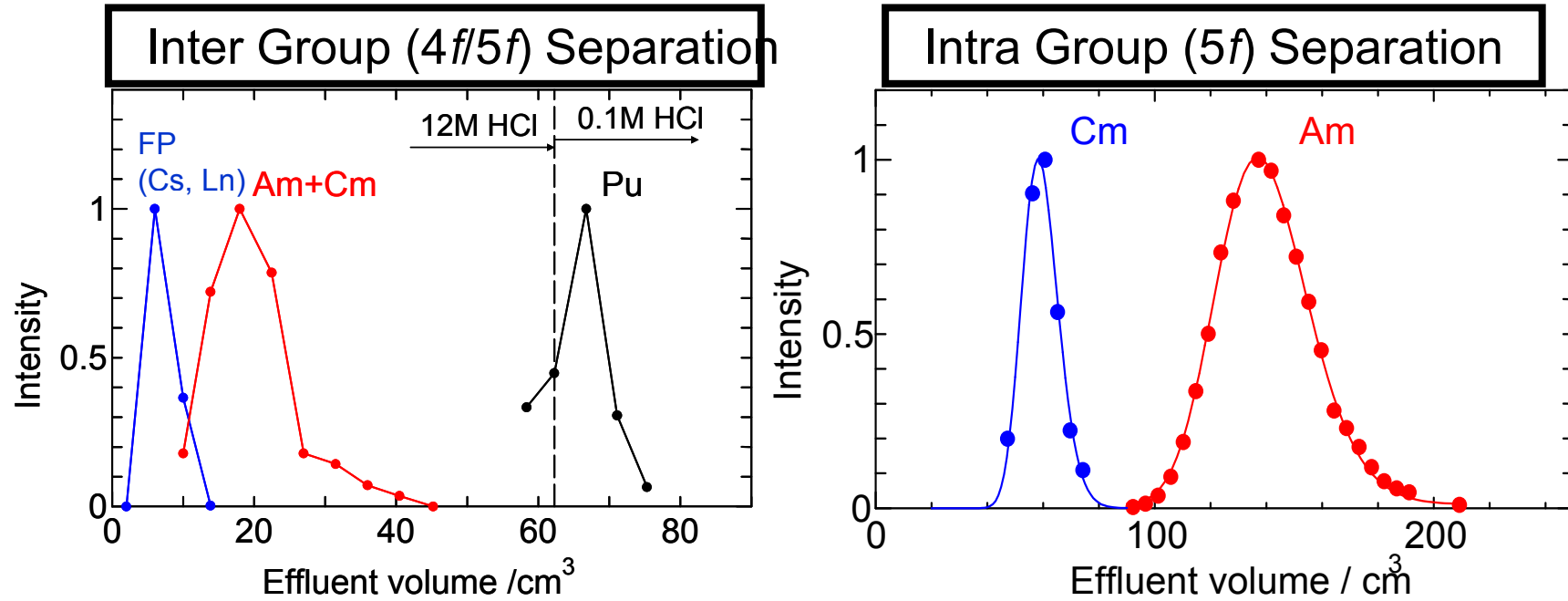
・HCl 中では, 分離係数は、イオン半径ではなく、むしろ 4f / 5f 元素のソフト性の差異に依存する。

実照射済み燃料を用いたアクティブ分離試験

* "Joyo" MOx SF (143.8 GWd/t, $2.23 \times 10^{27} (n/cm^2) (E \geq 0.1 \text{ MeV})$) : 1.27g/30ml



アクティブ試験結果



■ Perfect Removal of ^{106}Ru and ^{125}Sb

■ Recovery Rate of ^{241}Am Product

... possibly more than 99.9%

• >95% (for StepII + StepIII)

■ DF for ^{241}Am Product (MA/Ln Sptn.)

... far more than $1 \sim 2 \times 10^3$

• $\text{DF}^{155}\text{Eu} > 1.0 \times 10^5$

■ SF of ^{243}Cm for ^{241}Am Product (Am/Cm Sptn.)

• $\text{SF}_{\text{Cm/Am}} > 2.2 \times 10^3$

■ Purity of ^{241}Am Product (Cm/Am)

... far less than 1% (Ln / $\text{MA}_{\text{product}} < 5\%_{\text{mass}}$, CEA)

• $\text{Ln}(^{144}\text{Ce} + ^{155}\text{Eu}) / ^{241}\text{Am} < 0.3 \text{ ppm}$

• $^{243}\text{Cm} / ^{241}\text{Am} < 1 \text{ ppm}$

• $\gamma\text{FP}(^{106}\text{Ru} + ^{125}\text{Sb} + ^{137}\text{Cs}) / ^{241}\text{Am} < 1.8 \text{ ppm}$

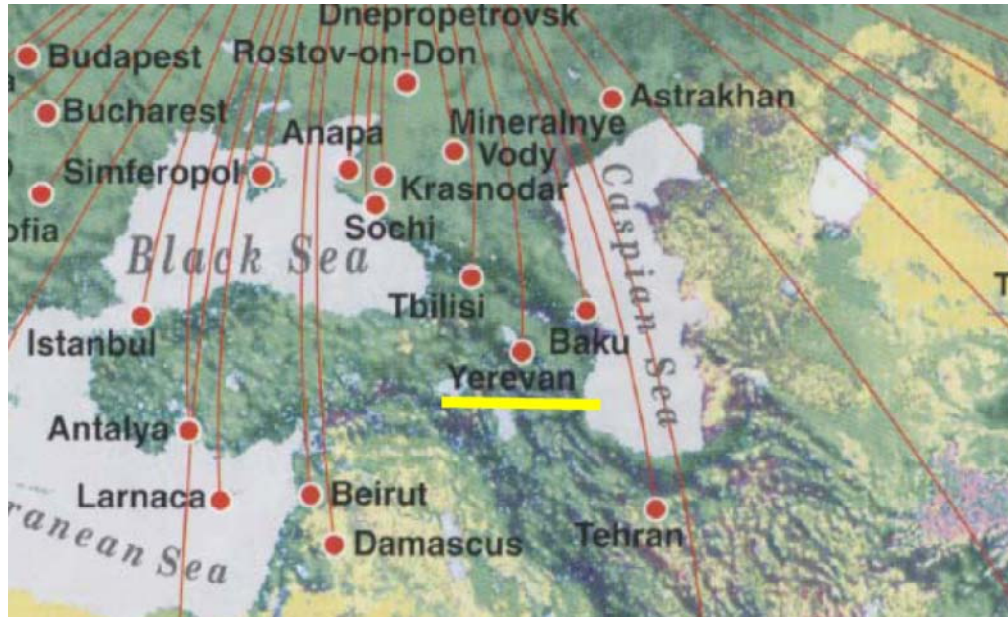
■ Purity of ^{243}Cm Product (Am/Cm)

... possibly less than 1%

• $^{241}\text{Am} / ^{243}\text{Cm} = 7.8 \times 10^3 \text{ ppm}$

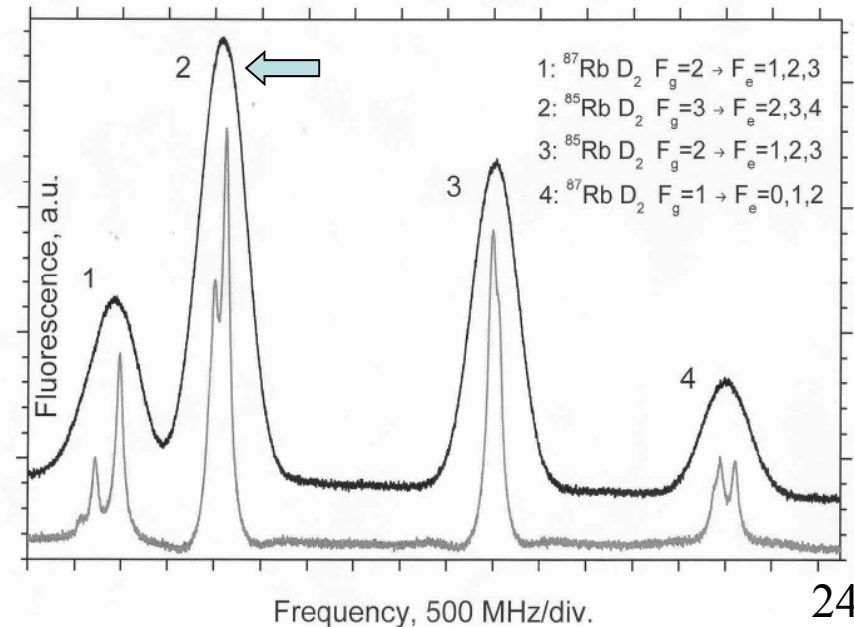
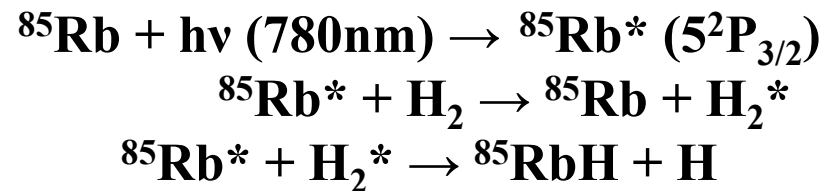
アルカリ金属 (Rb, Cs) の同位体分離研究

ISTC Collaboration with IP, Armenian Academy of Science (2002-2004)



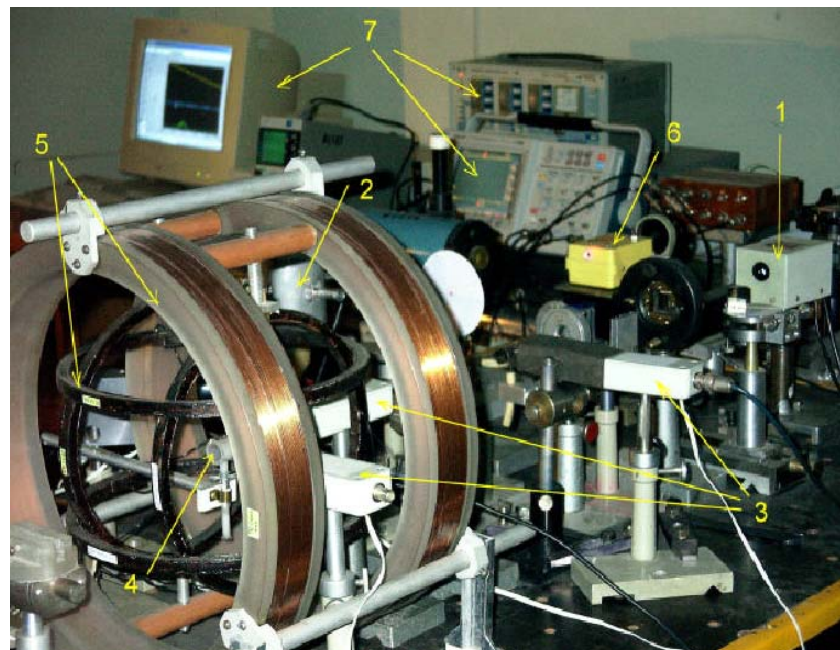
アルメニア原子力発電所 (PWR)

Laser-chemical Isotope (^{85}Rb) Separation - Mechanism



レーザー化学法 (LCIS) によるアルカリ金属の同位体分離

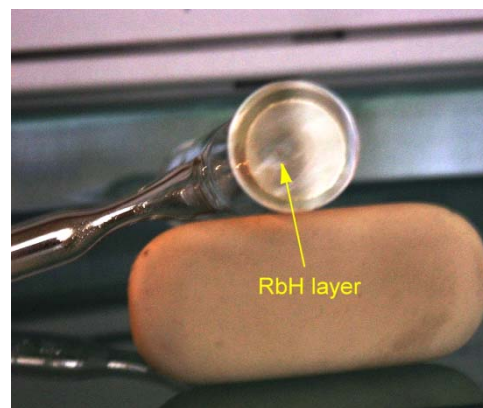
Item	Figure
Rb vapor density	5×10^{14} atom/cm ³
H ₂ density (converted into pressure)	5Torr
Cell temperature	220~240°C
Laser power, Irradiation time	50mW, 30min~2.5hour
⁸⁵ Rb ratio	72% (Before exp.) 98.4% (After two exps.)
⁸⁷ Rb ratio	28% (Before exp.) 1.6% (After two exps.)



Desk-top LCIS Equipments

1; 波長可変ダイオードレーザー、2; Rb(Cs)セル、3; 発光ダイオード、5; 交互交ヘルムフォルツコイル

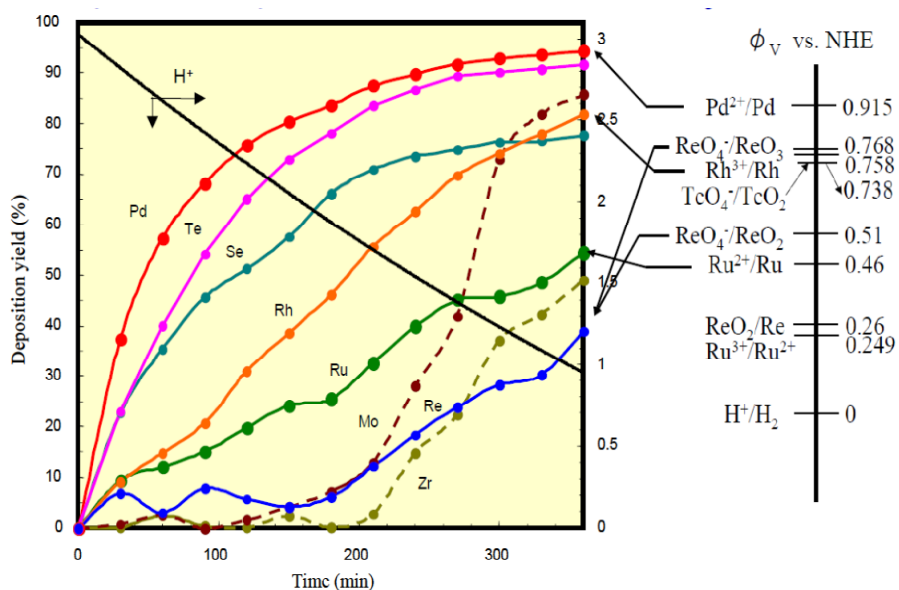
$$\frac{Y/(1-Y)}{X/(1-X)} \Rightarrow \text{Head Separation Factor was 2.99 (1st), and 23.9 (2nd)}$$



⁸⁵RbH Deposits at the Cell Window

触媒的電解採取法によるレアメタルの分離 UPD-enhanced CEE

●Basis of CEE (Catalytic Electrolytic Extraction) utilizing UPD (Under Potential Deposition)

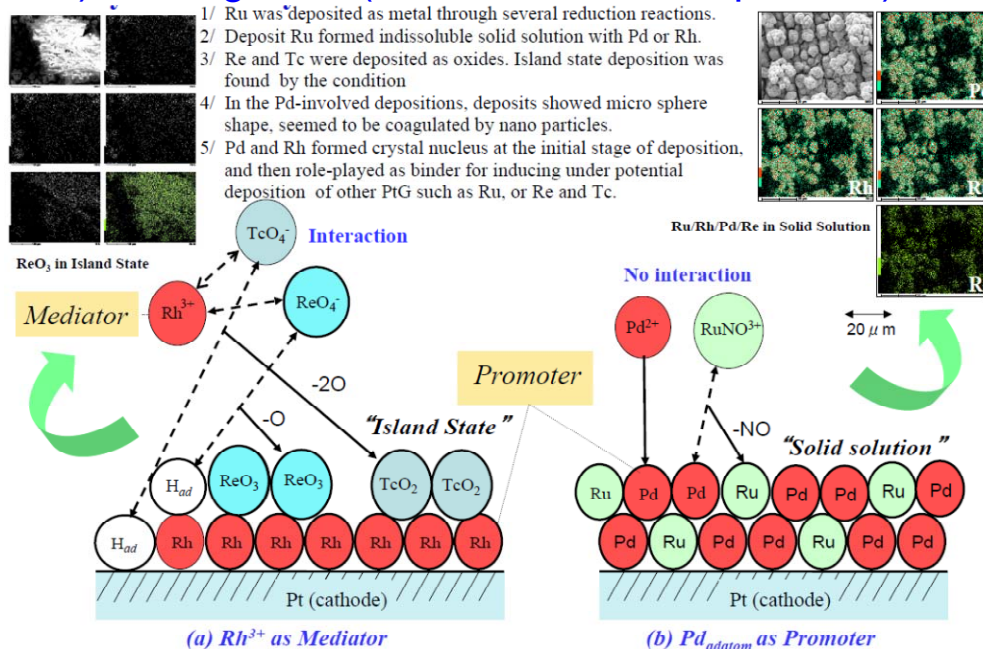
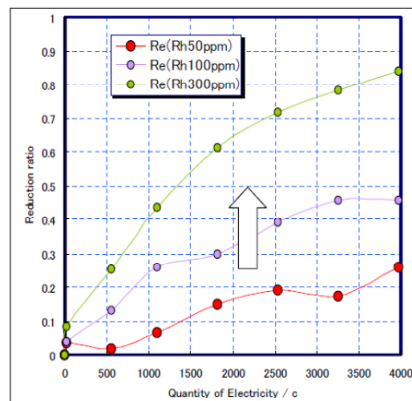
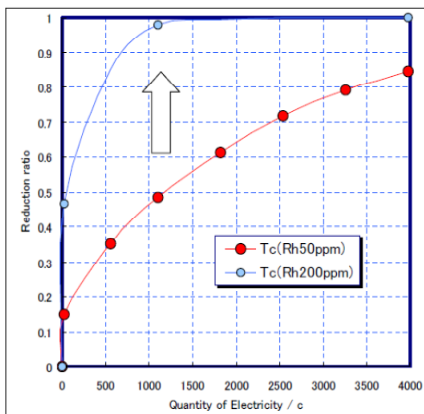


Separation of RMFP from Simulated HLLW

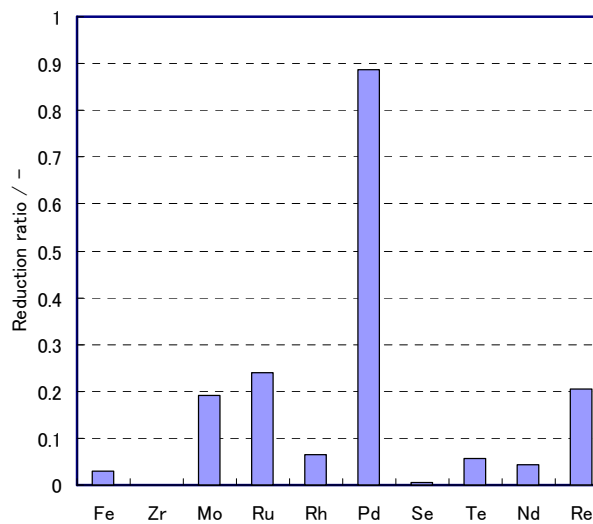
●Extension of CEE in HCl Media

CEE Conditions

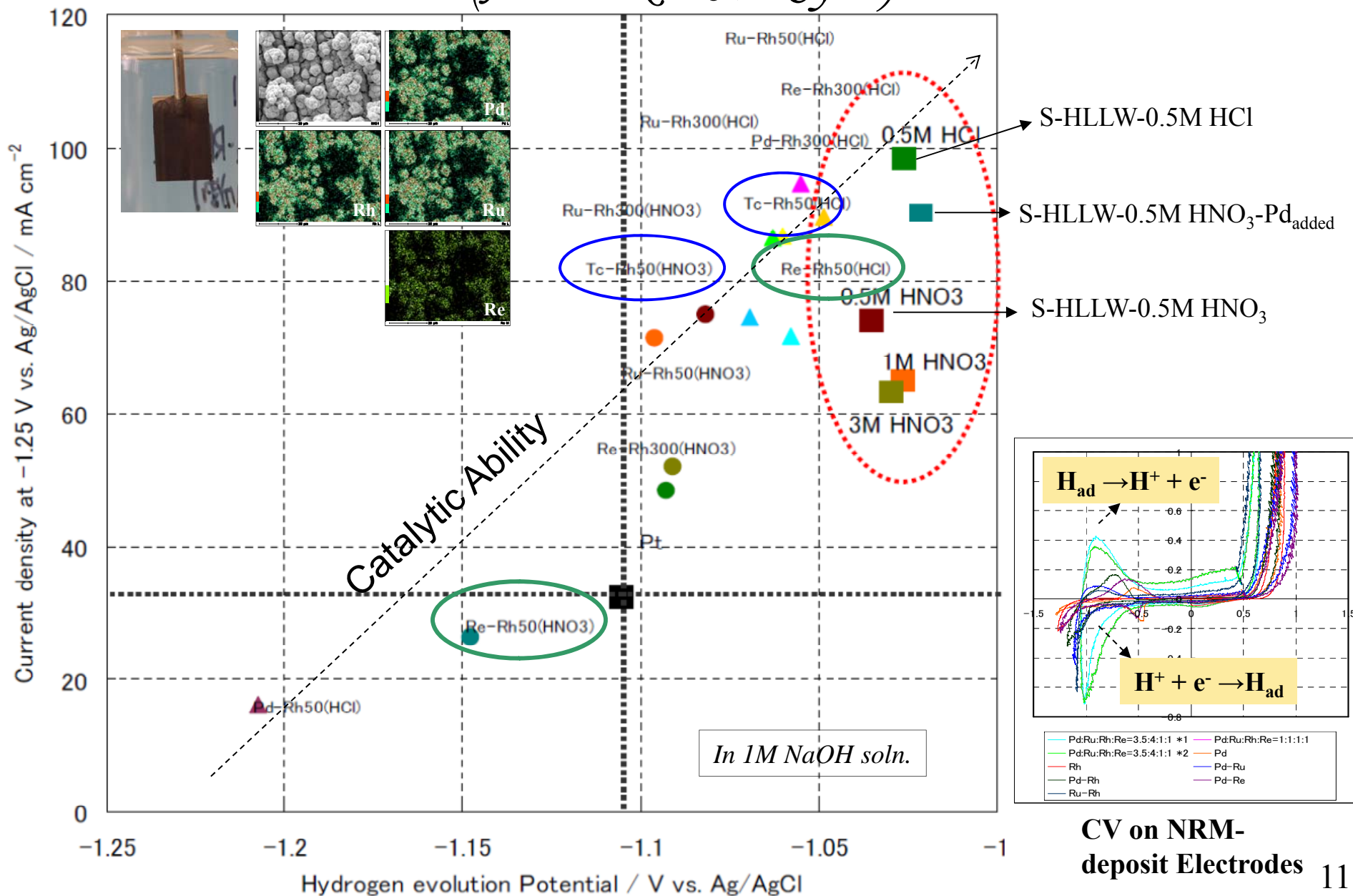
- Electrodes; Smooth Pt, Cathode (2cm²), Anode (8cm²), Ag/AgCl
- Catholyte; 0.5M HCl
- 50 °C
- Ic; 2.5mA/cm² (1hr) → 75mA/cm² (2hr) → 100 mA/cm² (4hr)



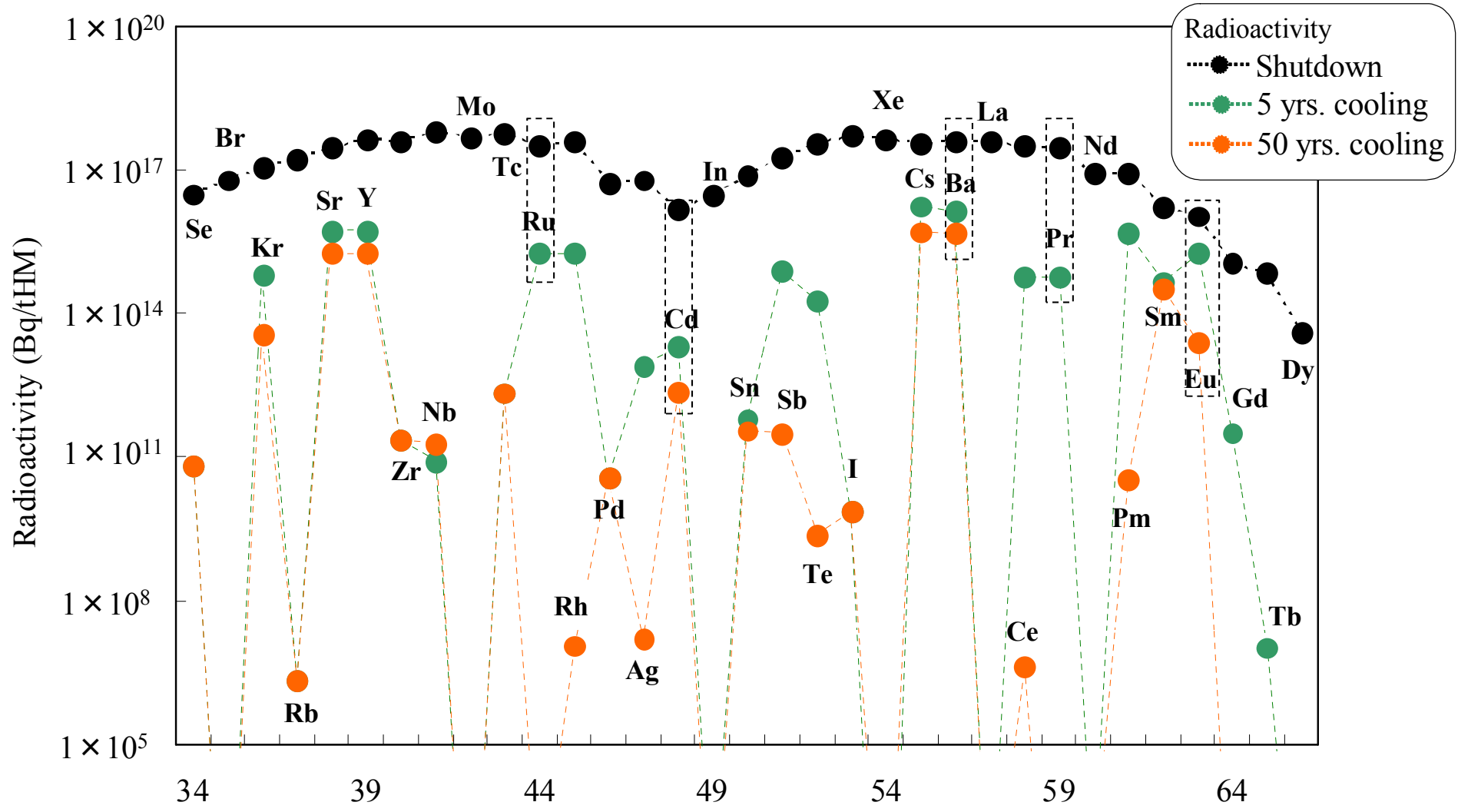
●Application of CEE in Simulated HLLW(HNO₃)



PGM, Tc, Re-Deposits as Catalysts for Electrolytic H₂ Production (Adv.-ORIENT Cycle)



Radioactivity of NRM As a Function of Cooling time for 50 years



FBR MOX, Inner core, 150GWd/t, cooled 5 years Atomic No.

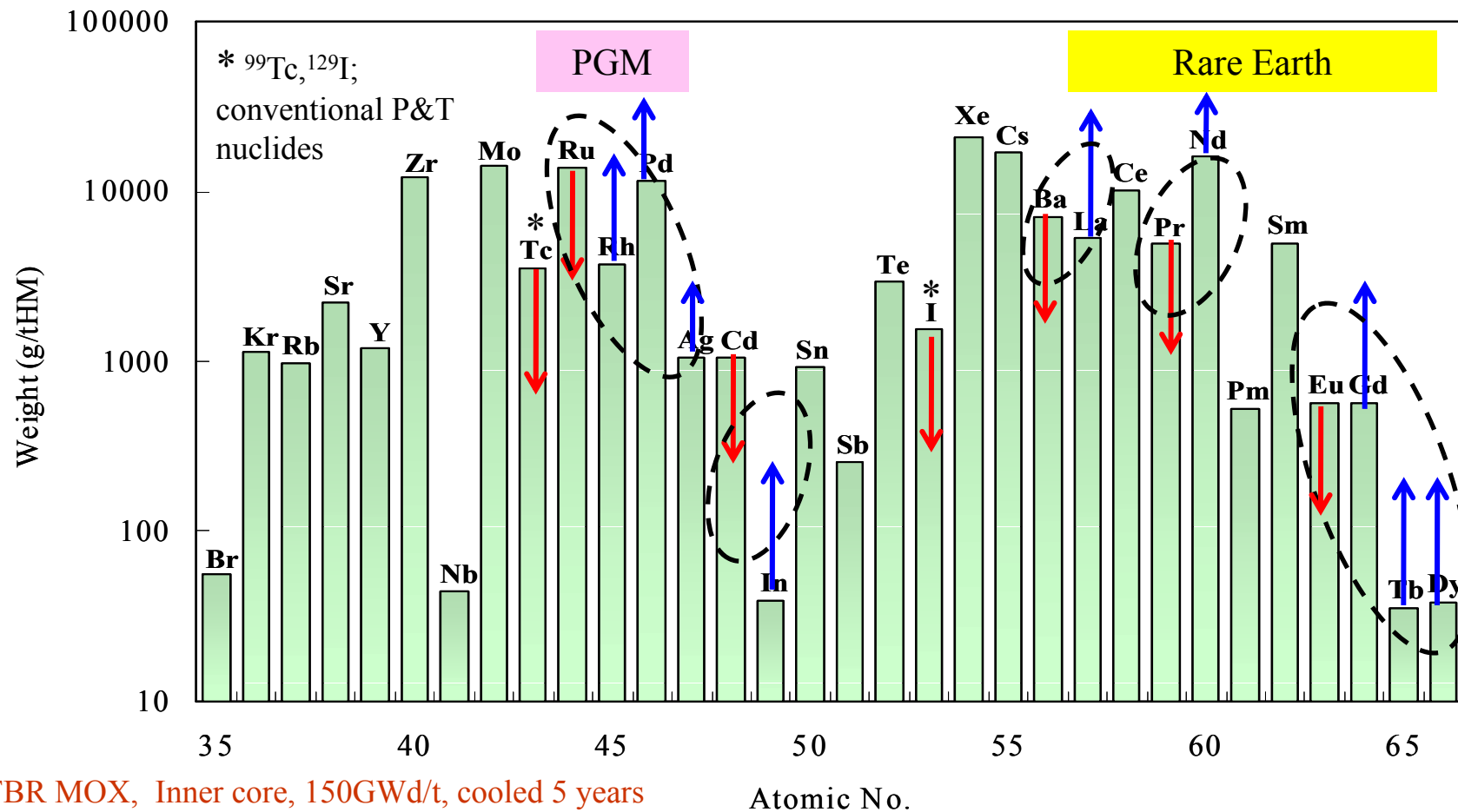
Après ORIENT (Nuclear Creation of Rare Metal)

① P&T: Transmutation of MA (^{241}Am , etc), LLFP (^{99}Tc , ^{129}I , etc)

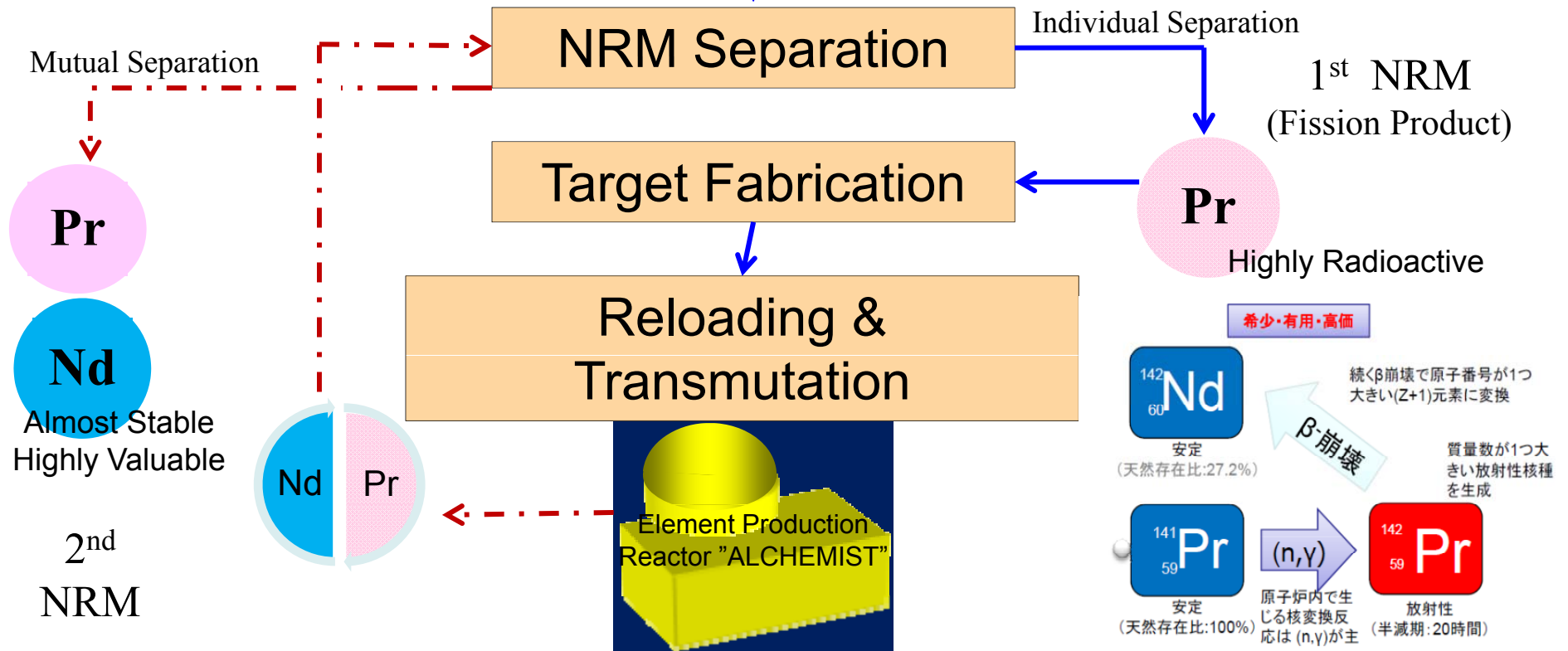
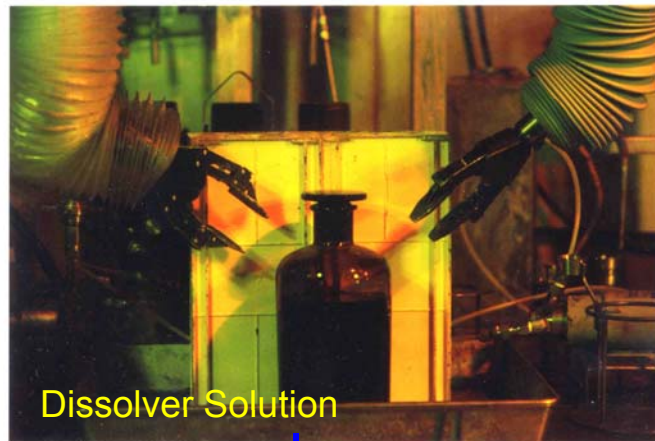
⇔ Decrease of HLW Radioactivity, Increase of Proliferation Resistance of Pu

② Positive P&T: Transmutation of Radioactive FPs to Highly Valuable Rare Metal

⇔ Element Strategy, Increase of Proliferation Resistance



From "Separation" to "Creation" (*Après ORIENT*)



Irradiation Conditions for Calculation

Composition of Actinides, etc

Economy type core (B.R. = 1.03)												
O/M ratio = 1.05												
Loaded fuel (kg/batch)												
Region	Pu content (%)	U235 (kg)	U238 (kg)	Pu238 (kg)	Pu239 (kg)	Pu240 (kg)	Pu241 (kg)	Pu242 (kg)	Np237 (kg)	Am241 (kg)	Am243 (kg)	Cm244 (kg)
Inner core	18.3	22.3	7407.5	19.4	955	566.5	75.9	68.9	8.8	35.3	17.6	17.7
Outer core	21.1	20.5	6802.5	21.3	1048	621.8	83.3	75.6	9.7	38.8	19.4	19.4
Axial Blanket	0	20.7	6885	0	0	0	0	0	0	0	0	0

Reloading: FBR Inner core
Irradiation: 800day/cycle * 4cycle
Inter-cycle inspection: 45.5days

Table Operation condition of Japanese fast reactor

	JOYO MK-II core	MONJU	Commercial Reactor
Thermal output (MWt)	100	714	3570
Power fraction	0.95	0.53	0.50
Number of subassembly	67	108	288
Lattice pitch (mm)	81.5	115.6	206.0
Stack length (mm)	550	930	1000
Active core volume (cc)	211974	1162393	10584188
Power density (W/cc)	448	326	168
Neutron flux ($n \cdot cm^{-2} \cdot s^{-1}$) ($E \geq 0.1$ MeV)	3.05×10^{15}	4.09×10^{15}	2.27×10^{15}

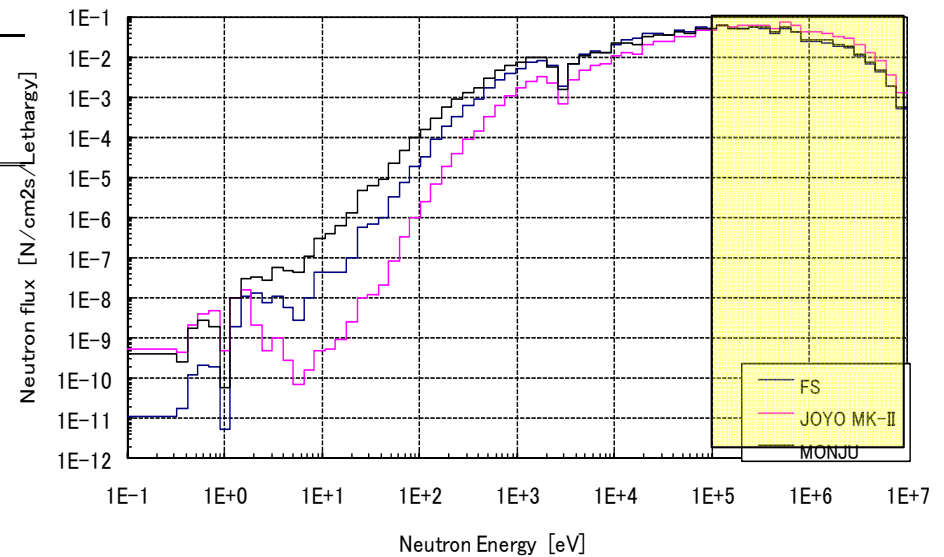
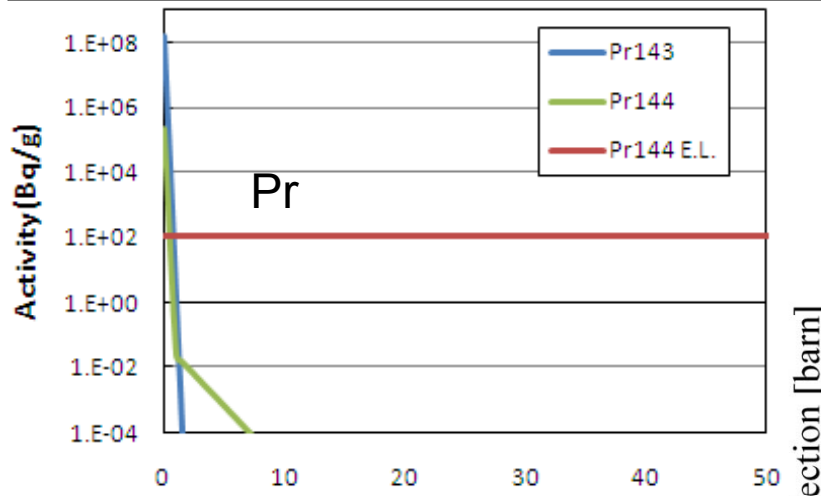


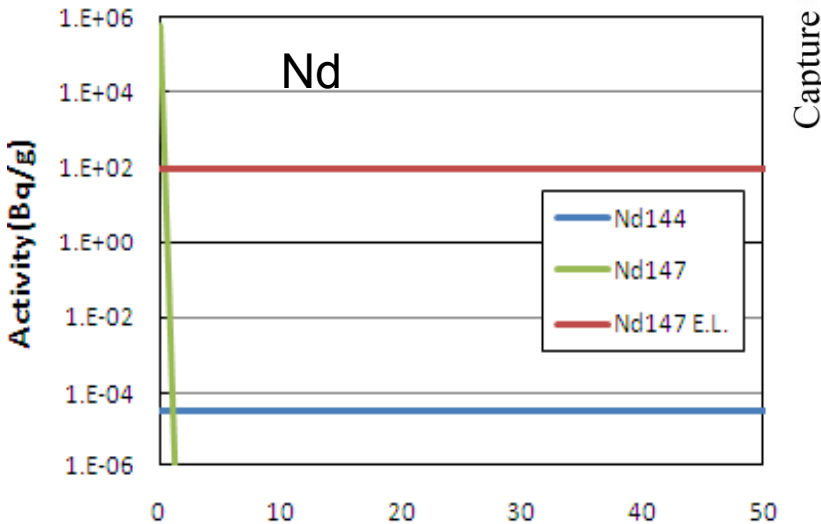
Fig. Neutron energy spectra at inner core

Transmutation of FP Pr

Radio-activities of re-irradiated FP Pr
(Pr, Nd products)



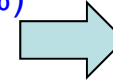
Cooling time(y)



Cooling time(y)

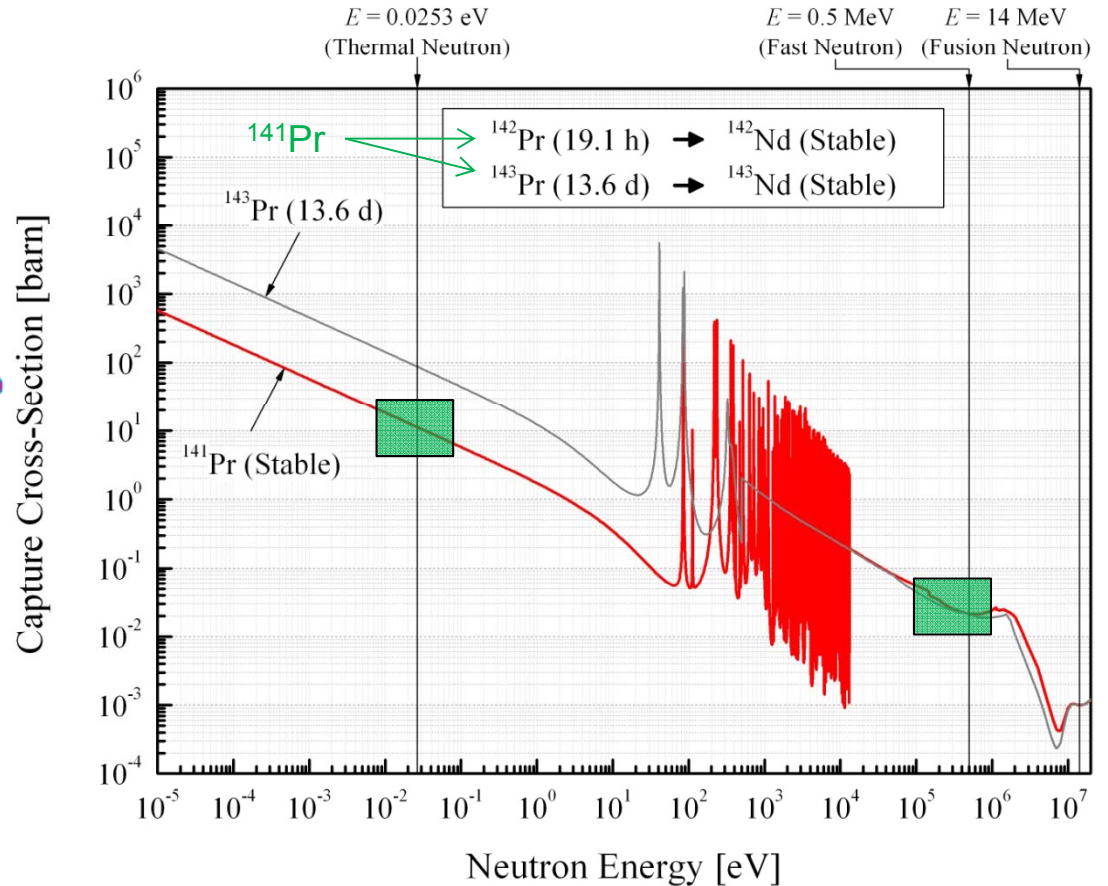
Loading Pr: 48.4kg (Pr-141:100%)

Shutdown: Pr 43.6kg, Nd 4.84kg



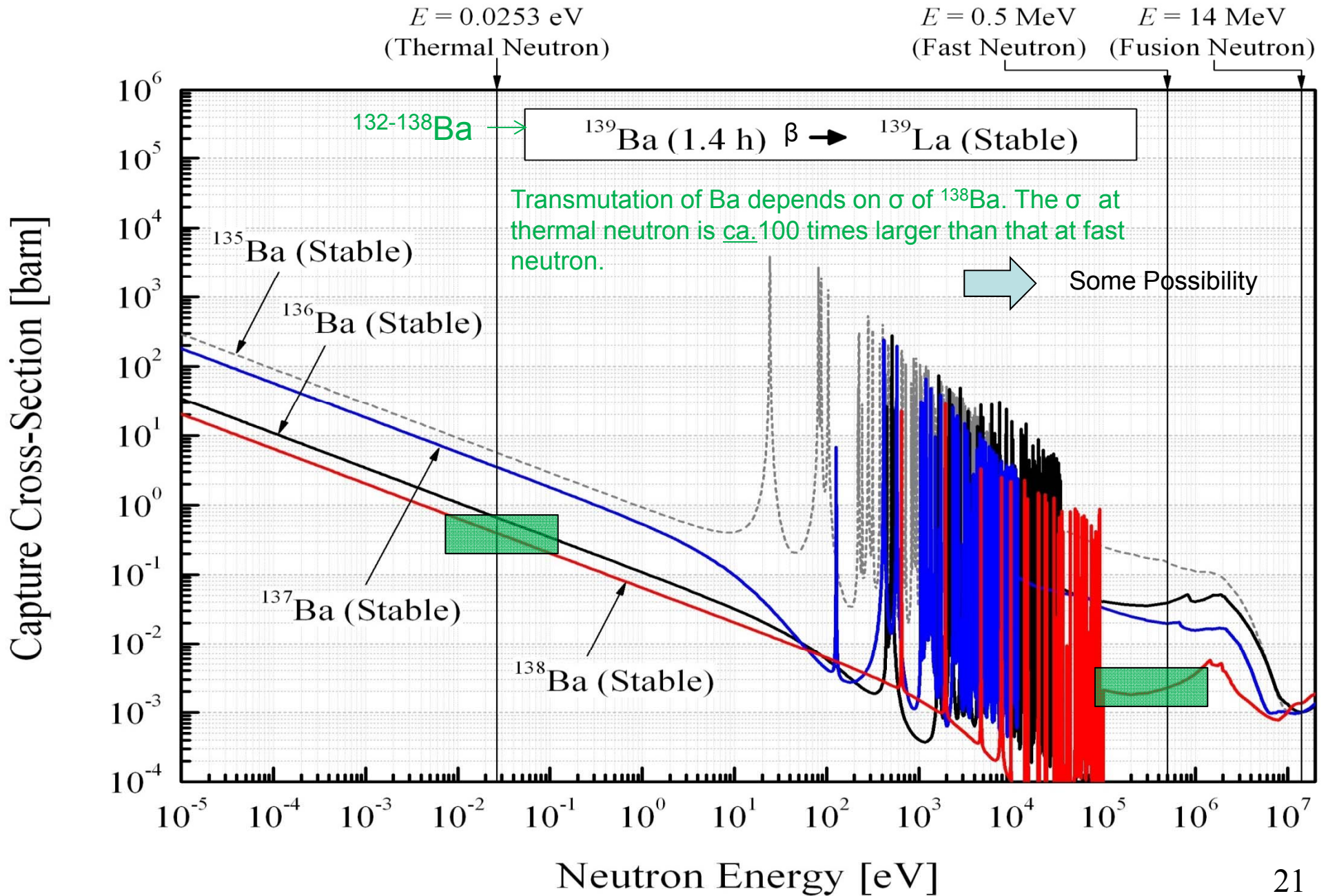
High Possibility

Transmutation ratio: 10.0%



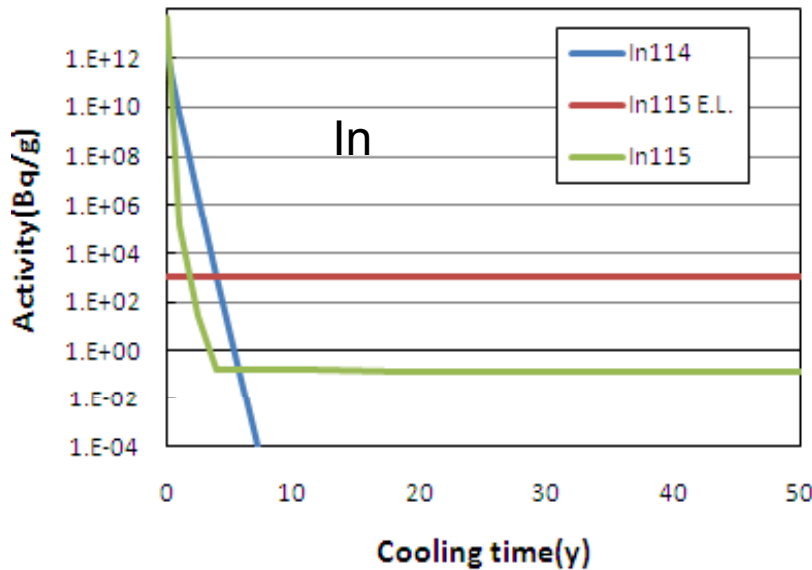
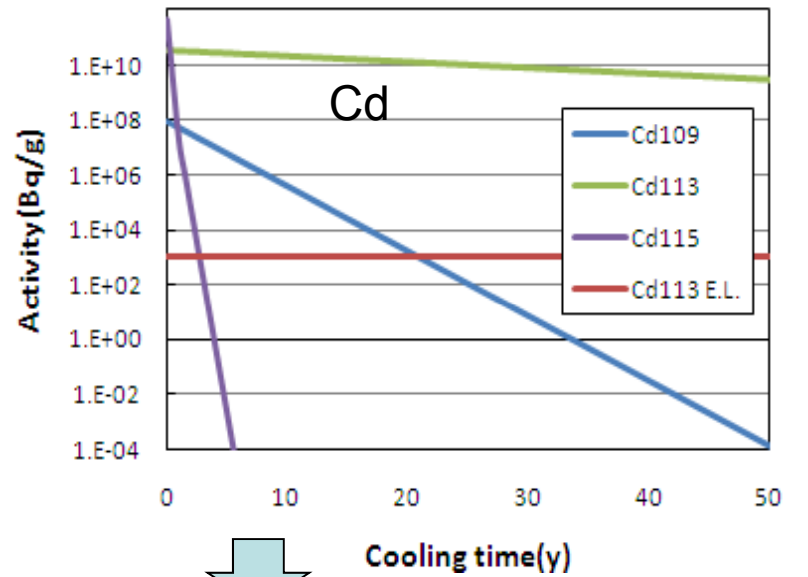
Transmutation of Pr depends on σ of ^{141}Pr . The σ at thermal neutron is ca. 1,000 times larger than that at fast neutron.

Transmutation of FP Ba

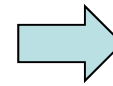


Transmutation of FP Cd

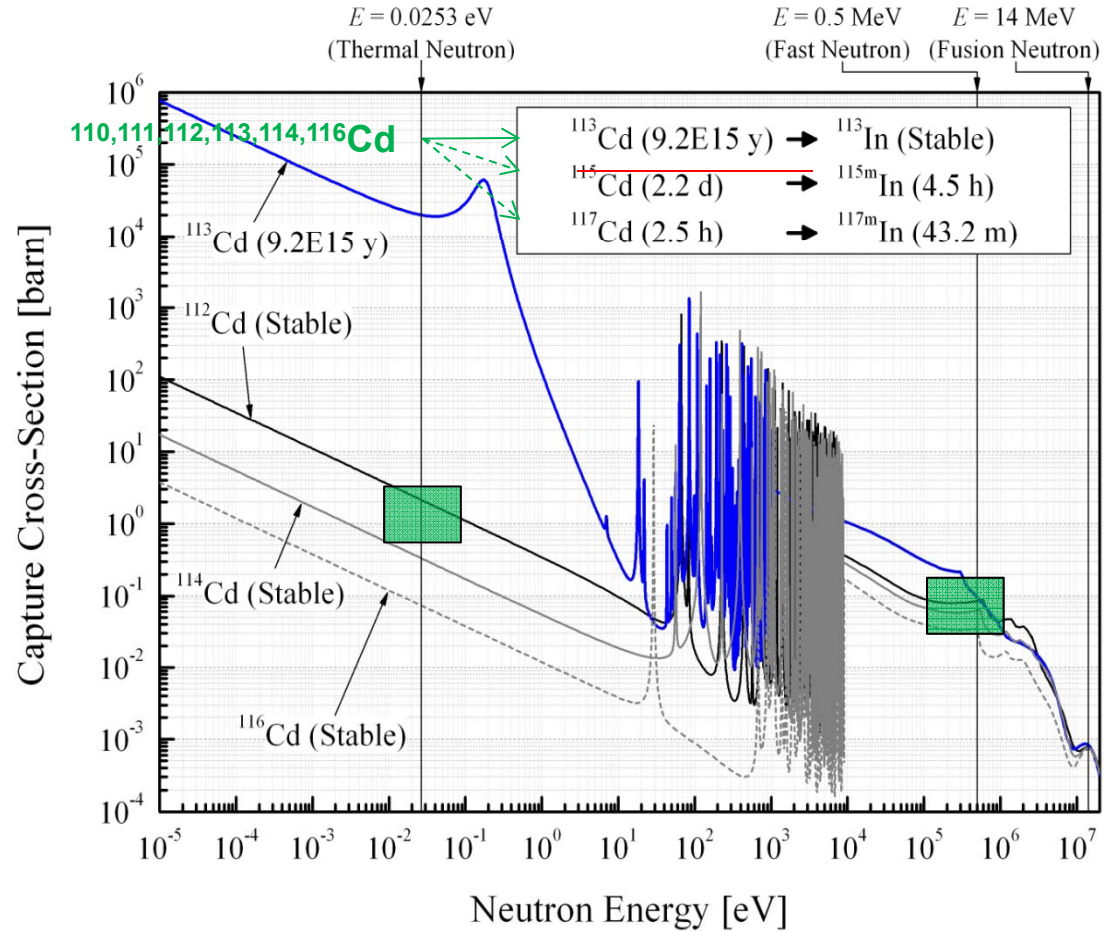
Radio-activities of Irradiated FP Cd
(Cd, In products)



Loading Cd: 10.3kg
Shutdown: Cd 10.0kg, In $8.1 \cdot 10^{-2}$ kg
Transmutation ratio: 0.8%



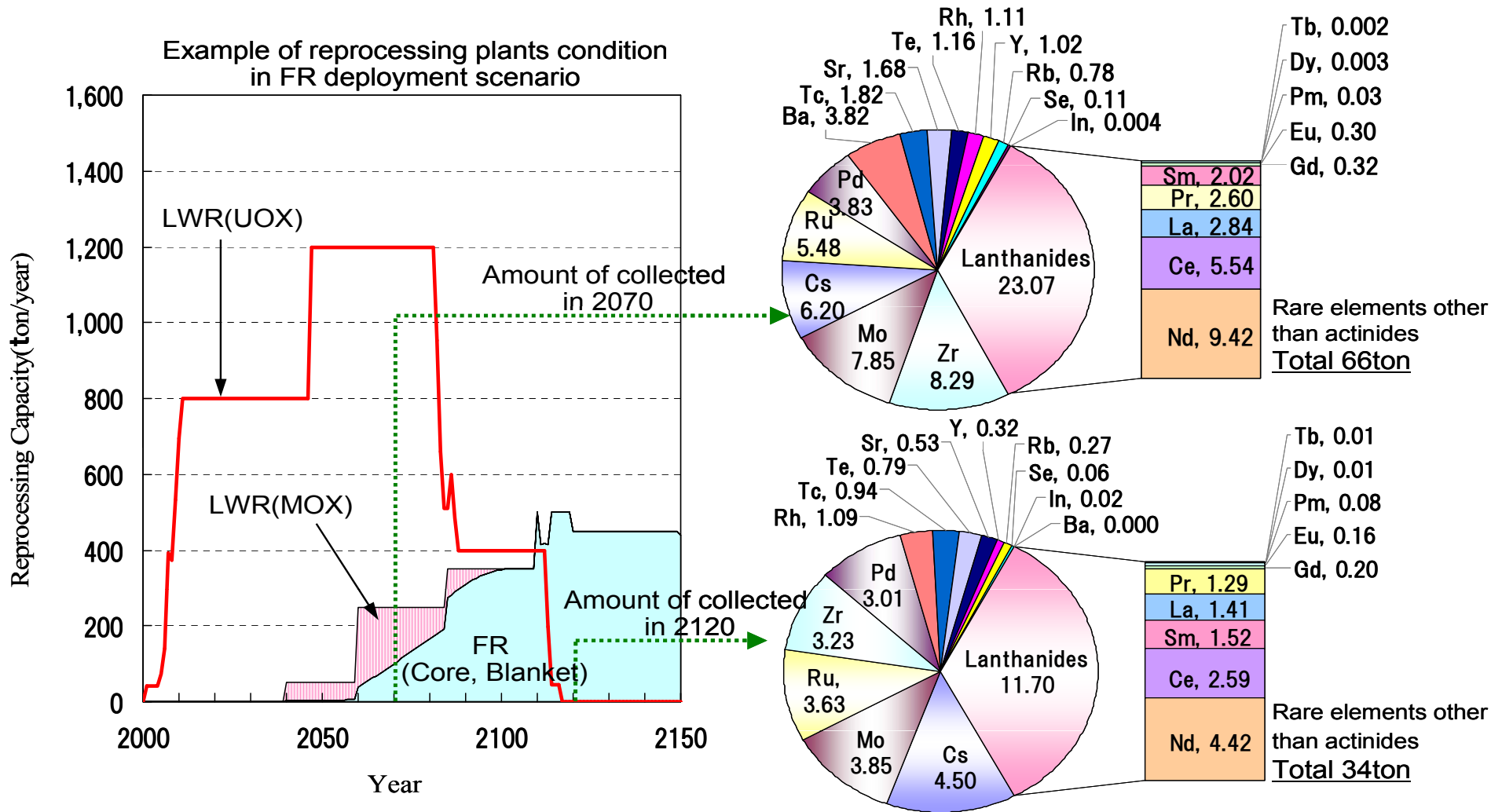
Less Possibility



Transmutation of Cd depends on σ of ^{112}Cd . However once ^{113}Cd created, transmutation to In will not proceed because of long-lived property of ^{113}Cd .

If NRM will be recovered in Adv.-ORIENT cycle - - - Japan's case

- National Demands of PGM in Japan (FY2006); Ru:3.7t, Rh:2.7t, Pd:50.6t
- In Estimating Nuclear Fuel Cycle Capacity in FY2070 in Japan, it can cover ca.100% of Ru, ca.40% of Rh and ca.7% of Pd against the national demands in FY2006.



* Previous Scenario until 3.11

*assume 99% as recovery ratio

下水から金 (Au) の回収



長野県(諏訪建設事務所)プレスリリース 平成 21 年(2009 年)1 月 28 日

諏訪湖流域下水道において汚泥焼却灰から『金』の回収を始めました。

諏訪湖流域下水道豊田終末処理場(愛称:クリーンレイク諏訪)の下水汚泥焼却灰等には金が含まれています。金が含まれている焼却灰は売却が可能であることから、平成20年10月から売却を始め、今月末に初めての売却収入があります。売却収入については、流域下水道の維持管理のために活用し、流域下水道を利用する皆様へのサービス向上を図ってまいります。金の回収の概要は、下記のとおりです。

実施前(今まで)

当処理場の汚泥焼却灰の中にはヒ素が多く含まれており、これの溶出を防ぐため溶融結晶化処理をして人工骨材として有効利用していますが、下記については骨材化が不可能なため、特別管理産業廃棄物として処分していました。

- 1 溶融飛灰(焼却灰を溶融結晶化する過程で発生するばいじん)
- 2 年1回の定期修繕に伴う溶融結晶化炉停止時の焼却灰
- 3 溶融過程で発生する不良スラグ



実施後(これから)

溶融飛灰等の金の含有量試験により、一定の含有が認められる場合、有価物として金属製錬会社へ売却します。

【平成20年10月分】

売却収益 約500万円(溶融飛灰 約500万円、焼却灰 0円)

排出量 溶融飛灰 1.4t 焼却灰 19t

金の含有量 溶融飛灰 約1,890g/t 焼却灰 約35g/t

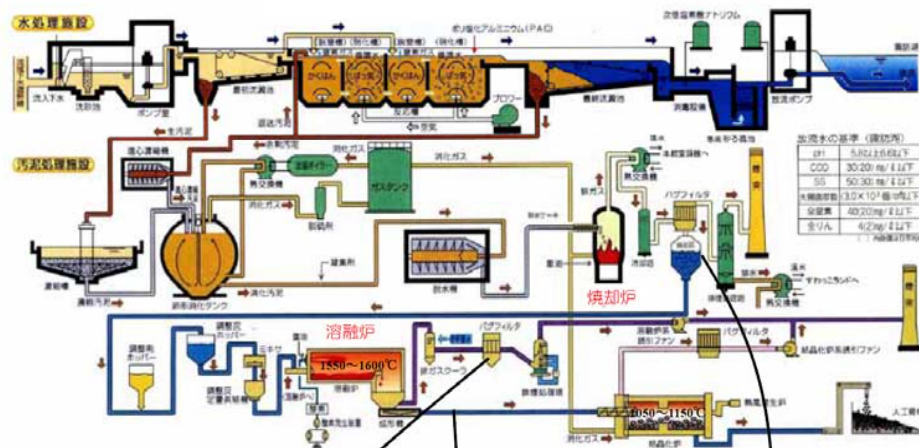
参考)平成20年度年間売却予想数量(金の市場価格及び含有量により変動します。)

溶融飛灰 5t

焼却灰 60t

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豊田終末処理場 処理フロー



金の回収の概要
平成19年10月から平成20年12月までに発生した溶融飛灰等を処分する中で、10月に搬出した溶融飛灰1.4tと焼却灰19tに金が含まれており、約500万円の売却収益がありました。残りの溶融飛灰等についても3月までに順次、売却収益額が確定します。

飛灰とは?
溶融炉で溶融処理する際、排ガスの中に飛んでいる灰で、バグフィルタで捕集されます。

処理場の運転経費
・処理場全体: 約14億3千万円
・溶融炉: 約1億7千万円

1890g x 5t x 約3500円(金)/g ≒ 3,300万円/年

原子炉で現代の錬金術を実現 *(Après ORIENT)*

- レアメタルは先端産業に直結した重要元素で、我国のGDPを制する。白金族、希土類及びレニウムなどは、資源が局在化しており、戦略物質として取り扱われる可能性が極めて高い。
- 核分裂反応はエネルギーの生産とともに31種を超えるレアメタルを創生する。使用済み燃料や高レベル放射性廃液を“廃棄物”ではなく“人工資源・鉱脈”として捉える発想（戦略）の転換と戦術の構築が必要である。
- 原子炉は新たな物質供給源（人工鉱山）として、資源小国である日本の資源確保戦略に寄与する可能性がある。
- 放射性レアメタルの核変換（ n, γ 反応）では、高価値レアメタル（例えば、 $Ru \rightarrow Rh$ ）の創成、あるいは低放射性元素（例えば、 $Eu \rightarrow Gd$ ）への転換、の可能性が認められた。
今後、全アクチニド及びFPの核変換特性を評価するとともに“元素創成”原子炉を視野においた検討を進める。



- Higher possibility can be expected by neutron capture cross sections (σ) even at the fast neutron energy spectrum (*i.e.*, $>0.1\text{MeV}$) conditions for $\Phi 2.27 \times 10^{15} \text{n/cm}^2/\text{s}$ and 8.8 years irradiation; $Ru \rightarrow Rh$ (t.r. 3.5%) / Pd (3.7%), $Pr \rightarrow Nd$ (10.0%), $Ba \rightarrow La$, $Eu \rightarrow Gd$ (64.8%), $Gd \rightarrow Tb$, $Tb \rightarrow Dy$. On the other hand, less possibility was recognized for; $Cd \rightarrow In$ (0.8%).
- Softer spectrum condition should be considered with expecting higher σ .
FBR-based feasibility calculation study will be made for target NRM subassembly with proper neutron moderators as $ZrH_{1.65}$, Be metal, etc.



Final Program

Nov. 30 - Dec. 3 2011, Shanghai, China



上海交通大学

Shanghai Jiao Tong University

東京工業大学

Tokyo Institute of Technology

日本国驻上海总领事馆



CONSULATE GENERAL OF JAPAN
SHANGHAI

东京工业大学原子炉工学研究所
小泽 正基
上海交通大学核科学与工程学院
书 悦周

关于“东京工业大学以及上海交通大学”

使用本馆名义作后援的许可

关于 2011 年 8 月 1 日申请的本案，现予以许可。

如该活动计划有所更改，须及时报告，请求许可的同时，请提出本活动开展后的结果报告书。

日本国驻上海总领事

泉 裕泰

2011 年 8 月 19 日

Summary report of 1st ASNFC2011

1. Summary

The 1st Academic Symposium on Nuclear Fuel Cycle (ASNFC2011, 11/30-12/3, 2011) was successfully held at Shanghai Jiao Tong University (SJTU) in China. This was the first joint conference of universities between China and Japan, focusing on basic chemistry and physics for nuclear fuel cycle. At the plenary session, a message was given by prof. Zhang Jie, president of SJTU (上海交通大学学長 張杰教授).

About **100** scientists from **25** universities and institutions attended, including about **40** young students from the both countries. Totally **42** high quality papers were presented, and very useful discussions were exchanged. In this time, three invited lectures were given from United States, Russia and Sweden.

Organizing committee thanks to the sponsors from China and Japan, and expresses the highest appreciation to the international advisory and the technical program committee members, especially for the local committee at SJTU.

The organization committee would like to propose Tokyo Institute of Technology, Japan as the next host organization university for the 2nd symposium, probably November of 2012.

2nd ASNFC; Nov. 2012, Tokyo Tech. (Japan) 東京工業大学
(Tentatively)

3rd ASNFC; Nov.2013, Peking Univ. (China) 北京大学

4th ASNFC ; Nov.2014, Tohoku Univ.(Japan) 東北大学
以降、四川大 (中国)、清华大 (中国) . . .

The committee is also discussing to expand academic topics and universities to in-Asia (Korea, India, etc.) and also ex-Asia (United States, Russia and Europe),

2. Award announcement

The Best Presentation Award (student) for 4 students (CHN 2, JPN 2).

Kei Yamanishi (Tohoku Univ.) 東北大学

Masahiko Nakase (Tokyo Institute of Technology) 東京工業大学

Ying Dai (Zhejiang University) 浙江大学

Fuwan Zhai(Sichuan University) 四川大学

第2回日中大学間核燃料サイクル学術討論会
(2nd ASNFC 2012)
2012年秋 於東工大

百家争鸣 一律平等の議論！
熱烈歓迎 来たれ若者

