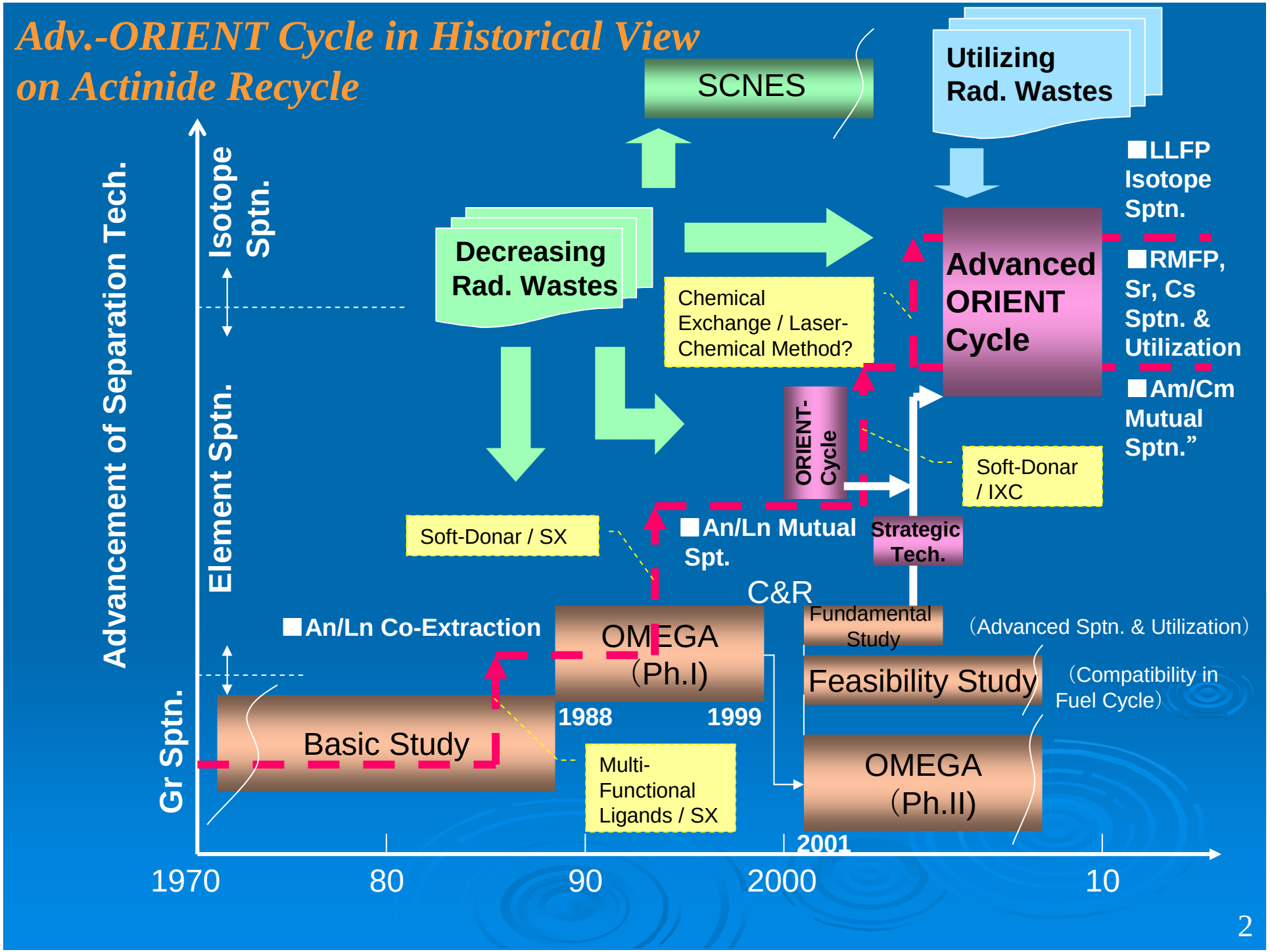
ICENES 2007
13th International Conference on
Emerging Nuclear Energy Systems
3-8 June 2007 Istanbul/Turkey
Parallel Session 10A: Fission Reactors V

Advanced ORIENT Cycle for Strategic Separation, Transmutation and Utilization of Nuclides in the Nuclear Fuel Cycle, Focusing on Electrochemical Separation and Utilization of RMFP

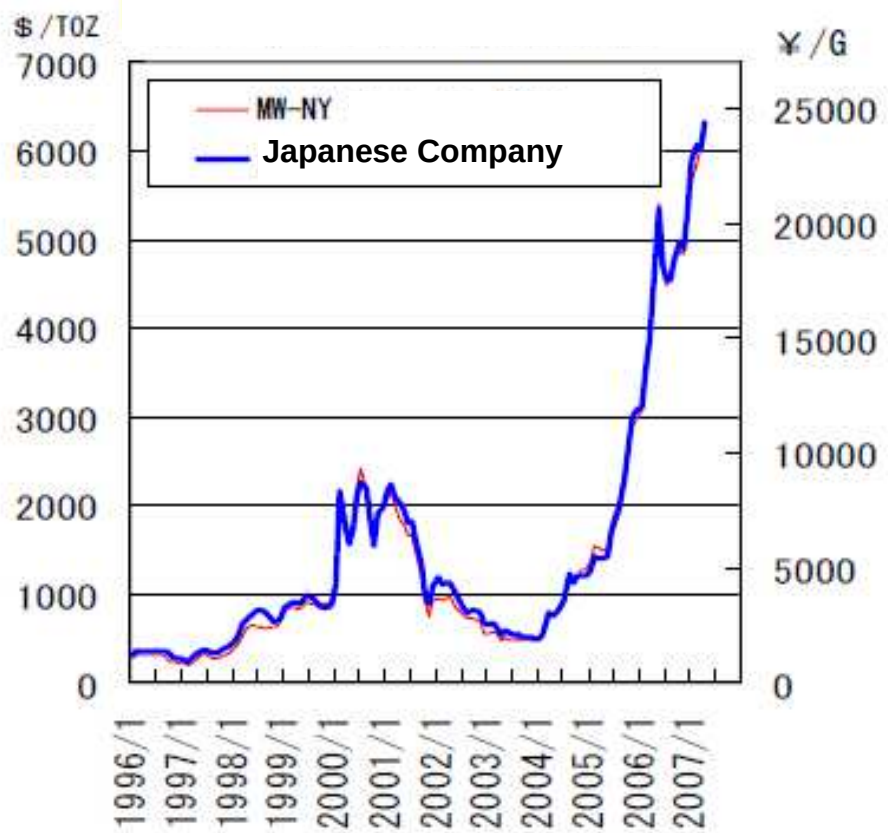
Masaki Ozawa[1] and Reiko Fujita[2]

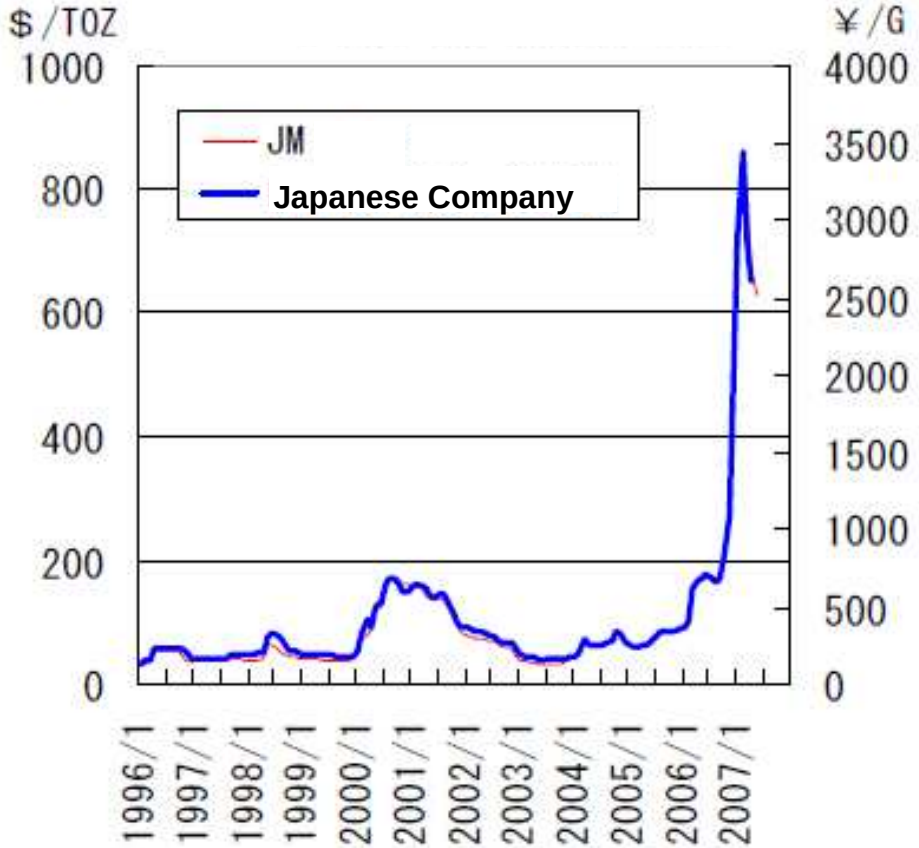
†1 Japan Atomic Energy Agency, Japan
†2 Toshiba Corporation, Japan

Adv.-ORIENT Cycle in Historical View on Actinide Recycle



Market Prices Shift on PtG Elements





Rh : 24,700

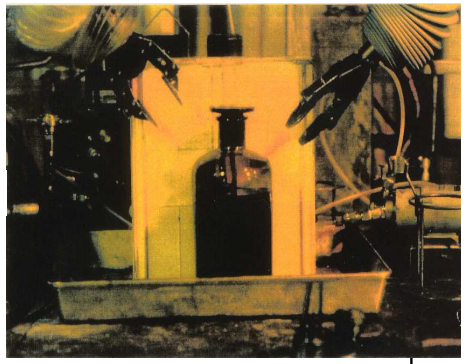
Ru : 2,220

Ir: 1880

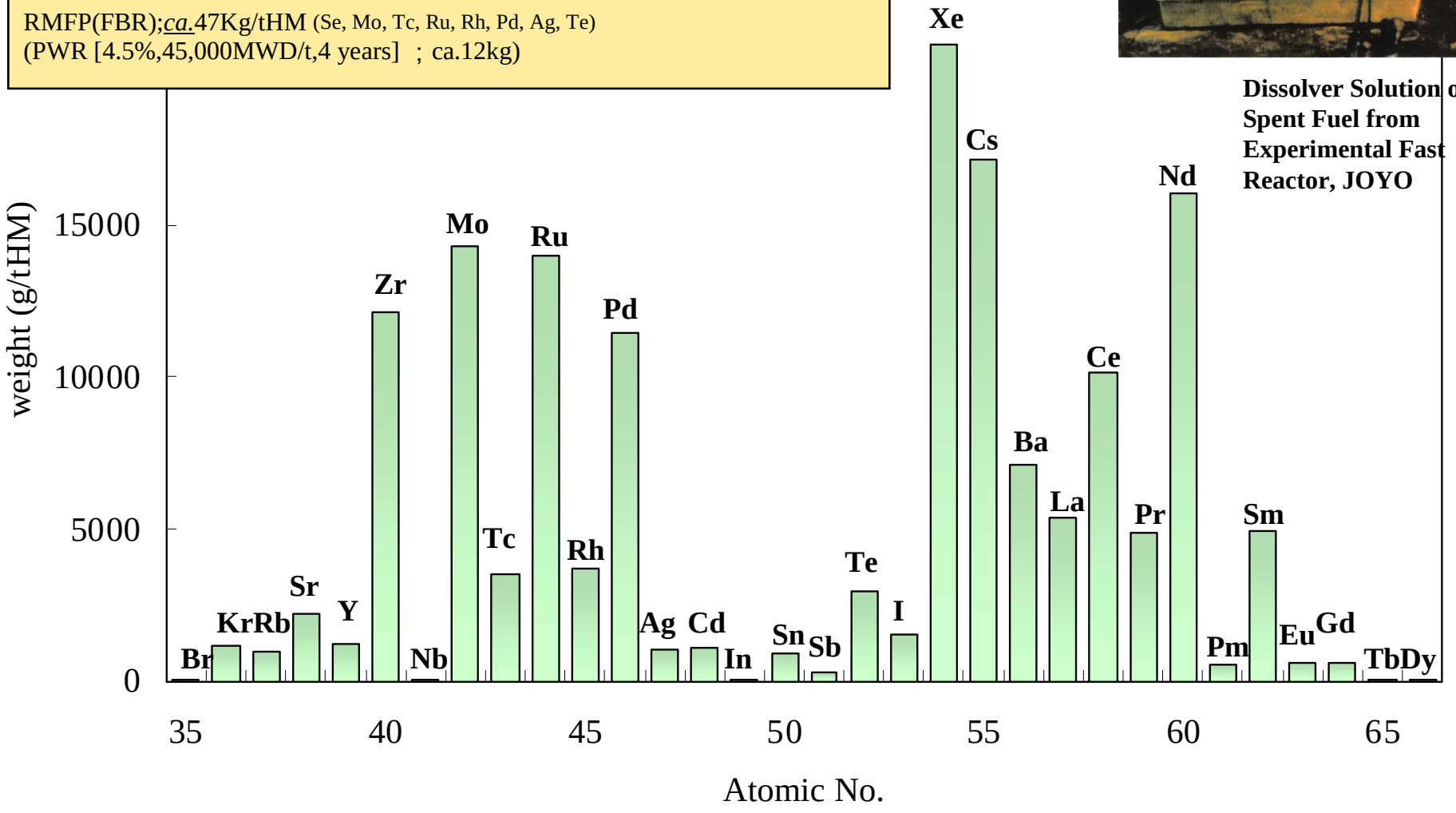
Spent Fuel, as Artificial Ore

Rare metal	Ru	Ph	Pd	Tc	Te	Se	Note
Weight (Kg/HMt)	13	3.8	12	3.5	3	0.2	FBR, 150GWd/t, 5 years cooling

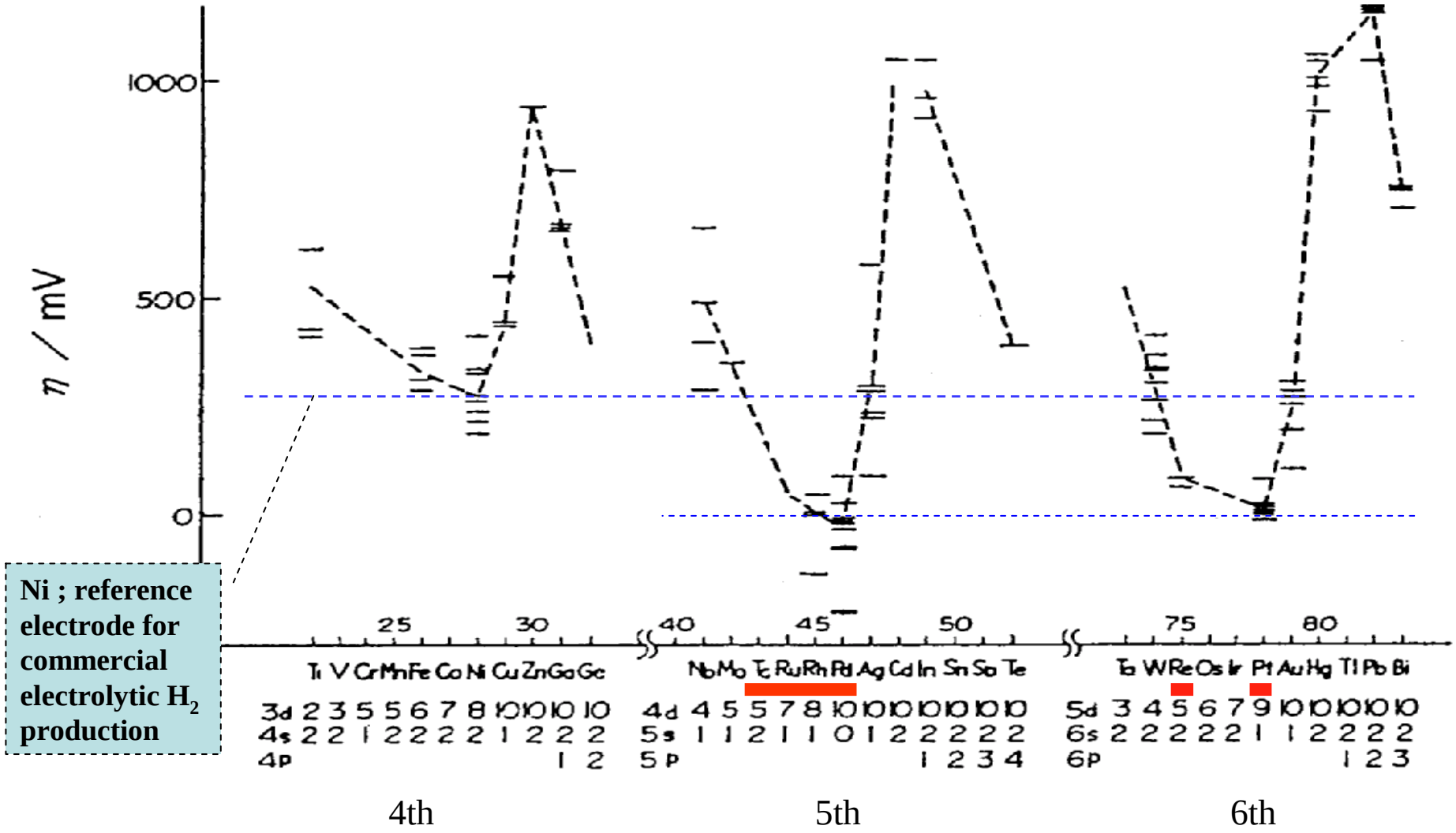
RMFP(FBR);*ca.*47Kg/tHM (Se, Mo, Tc, Ru, Rh, Pd, Ag, Te)
(PWR [4.5%,45,000MWD/t,4 years] ; ca.12kg)



Dissolver Solution of Spent Fuel from Experimental Fast Reactor, JOYO

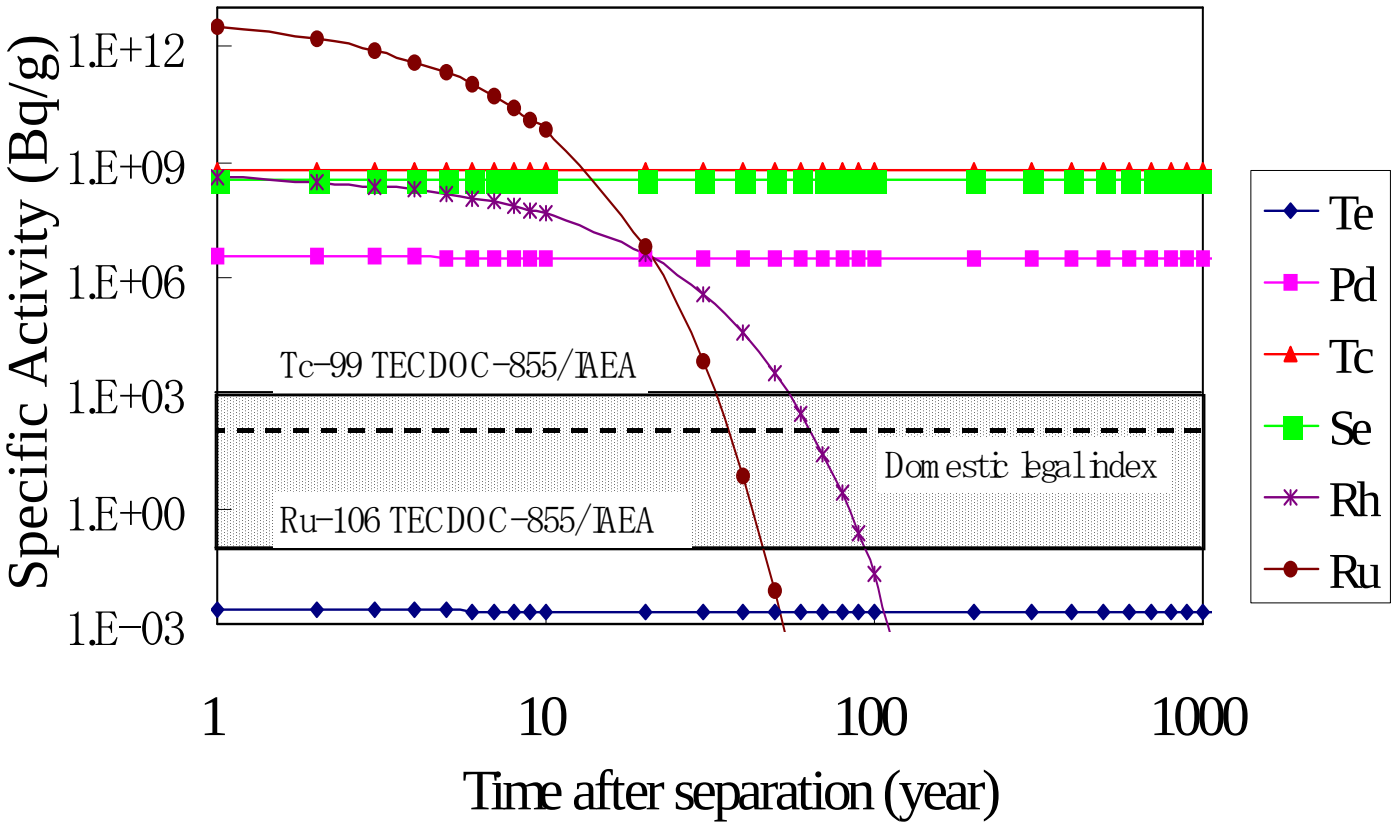


How to Utilize RMFP ?



Periodicity of Hydrogen Overpotential (Acidic Solution, Ic :1mA/cm^2) (Ref.) H.Kita, et al. Denkikagaku, 38,17 (1970)

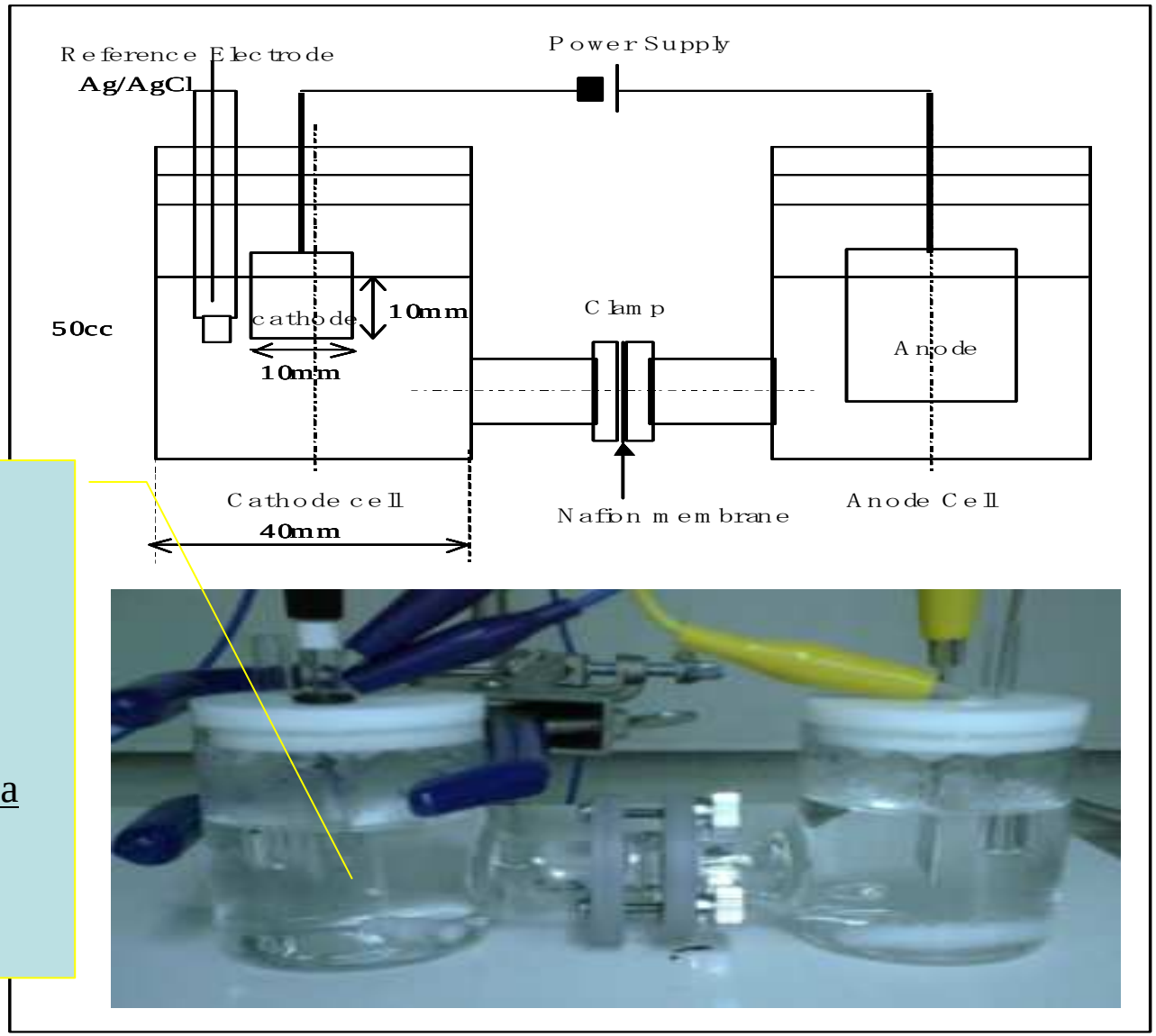
How much Radioactivity of RMFP at Use?



Time Dependency of Specific Activities of RMFP Separated from the FBR Spent Fuels cooled for 4 years

"Clearance levels" are proposed by IAEA"TECDOC-855" from 10^{-1} to 10^{3} Bq/g, for instance, 0.1≦Ru-106 <10、 100≦Tc-99<1000 (as for reference, 74 Bq/g , Japan domestic legal index).

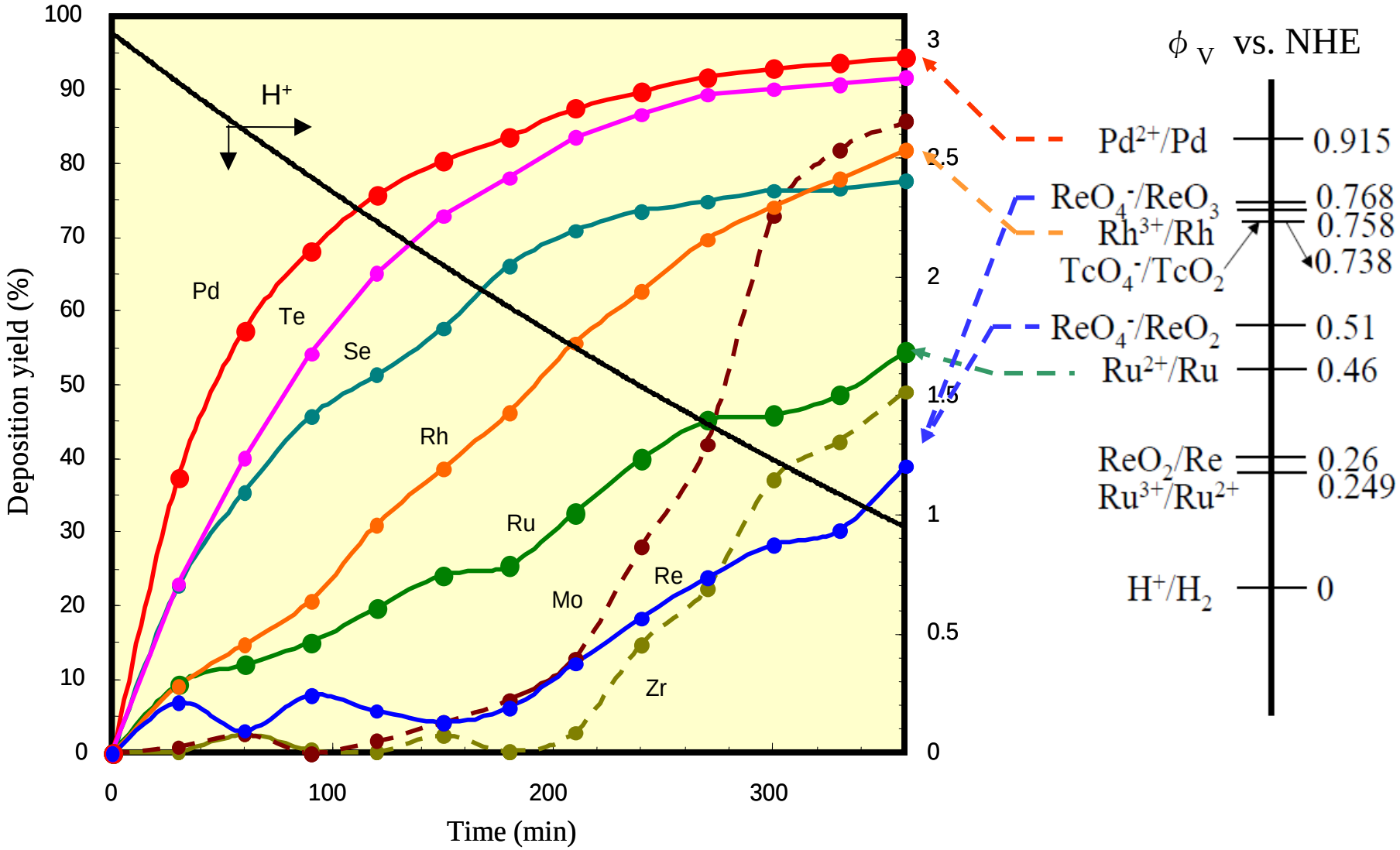
How to Separate RMFP ?



Catholytes ; S・HLLW for electrolytic extraction tests, NaOH or Sea Water in the case of H_2 production tests

Electrolytic Cell for Separation of RMFP from Simulated HLLW and for Generation of Hydrogen by Electrolysis

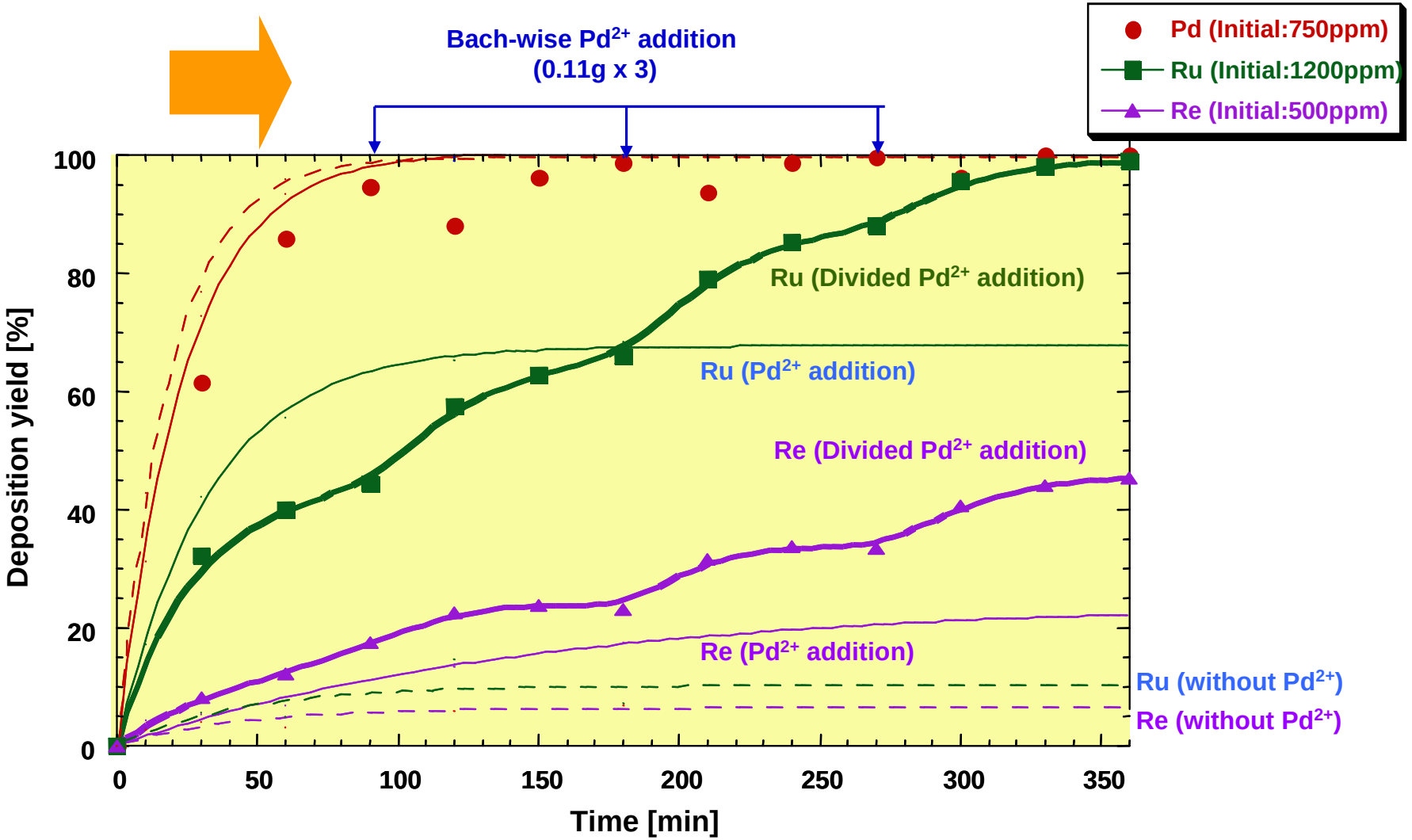
Fundamental Electro-deposition Behavior of RMFP in HLLW



Separation of RMFP from Simulated HLLW

Galvanostatic Electrolysis ; 500mA/cm^2 (Cathode), Room temp. Cathode; Pt-Ti, 20cm^2, S/V: 1/15cm^{-1}, Pd^{2+} Addition; Continuously (2.53gPd^{2+}/hr), Pd$_{added}$/Ru=3.6,

How to Improve the Deposition of Ru, Re and Tc?



Electrolytic Extraction of $RuNO^{3+}$ and ReO_4^-

***CEE* Effect by Addition of Pd^{2+} Electrolysis condition ;**

H^+ : 2.5M, Temp. : 50℃, CD : 500mA/cm^2, Pt cathode

Systematic Electrolysis Runs

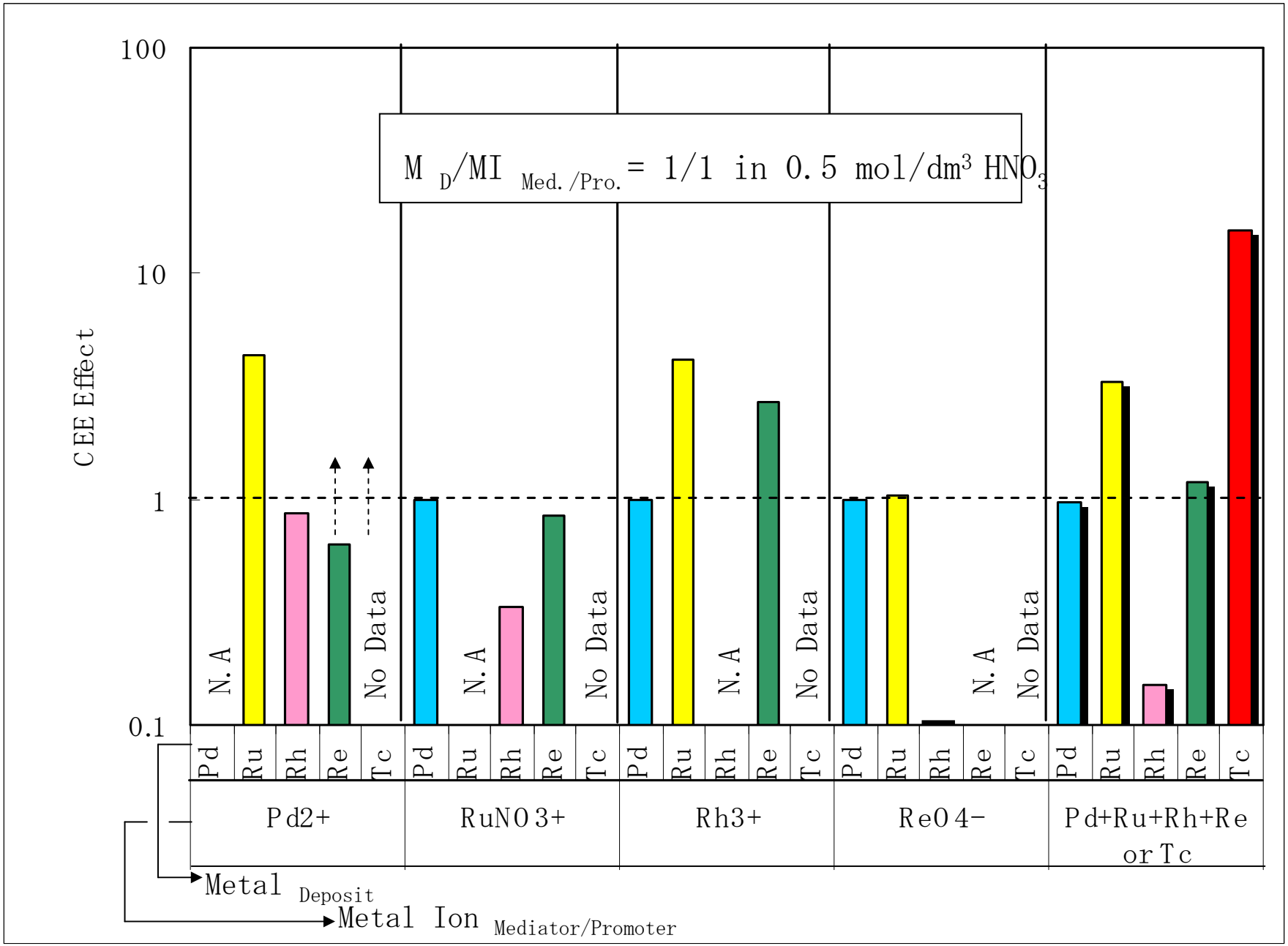
No.	System	Reduction ratio (%)				
		Pd	Ru	Rh	Re	Tc
1	Pd	>99	—	—	—	—
2	Ru	—	14	—	—	—
3	Rh	—	—	>99	—	—
4	Re	—	—	—	16	—
5	Tc	—	—	—	—	1.7
6	Pd-Ru	99.3	60.9	—	—	—
7	Pd-Rh	99	—	84.7	—	—
8	Pd-Re	99.4	—	—	10	—
9	Ru-Rh	—	58.2	32.5	—	—
10	Ru-Re	—	14.5	—	13.5	—
11	Rh-Re	—	—	10	43	—
12	Pd-Ru-Rh-Re(1:1:1:1)	95.7	46	14.5	19	—
13	Pd-Ru-Rh-Re(3.5:4:1:1)	99	11.8	2.1	33.4	—
14	Pd-Ru-Rh-Re(3.5:4:1:1)*1	94.7	16.5	26.6	55.3	—
15	Pd-Ru-Rh-Re(1:1:1:1)	90	35	15	20	—
16	Pd-Ru-Rh-Re(3:1:1:1)*1	50	32	7	12	—
17	Pd-Ru-Rh-Re(5:1:1:1)*1	60.9	54.8	19.8	12.8	—
18	Pd-Ru-Rh-Re(1:3:1:1)	72	13	0	5	—
19	Pd-Ru-Rh-Re(1:5:1:1)	77	10.7	0	0	—
20	Pd-Ru-Rh-Re(1:3:1:1)	85.3	6.8	13.6	9.9	—
21	Pd-Ru-Rh-Re(1:1:3:1)	96.1	39.7	7.7	16.2	—
22	Pd-Ru-Rh-Re(1:1:5:1)	93.8	41.7	6.0	9.9	—
23	Pd-Ru-Rh-Re(1:1:1:3)	93.2	36.5	6.1	9.8	—
24	Pd-Ru-Rh-Re(1:1:1:5)	93.2	32.9	3.7	7.9	—
25	Pd-Ru-Rh-Tc(1:1:1:0.5)	86.3	41.1	13.4	—	26.5

*1 Pd divided addition

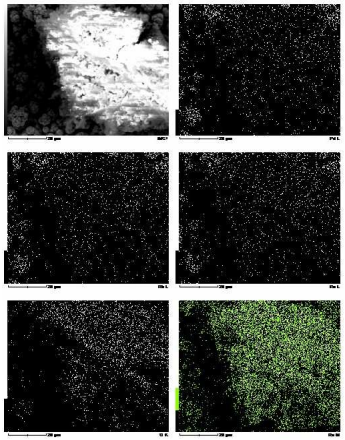
CEE conditions;

- Smooth Pt cathode(2cm^2)
- Catholyte: 0.5MHNO_3
- 50 ℃
- Ic （Run12-25): 2.5mA /cm^2 (1hr) → 25(2hr) →50(2hr) →100 (2hr)

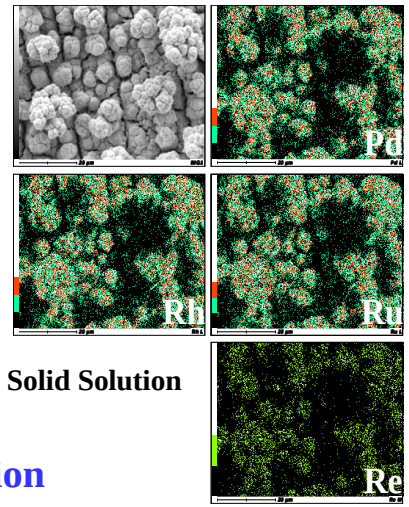
Acceleration of Deposit ; CEE (Catalytic Electrolytic Extraction) Effect



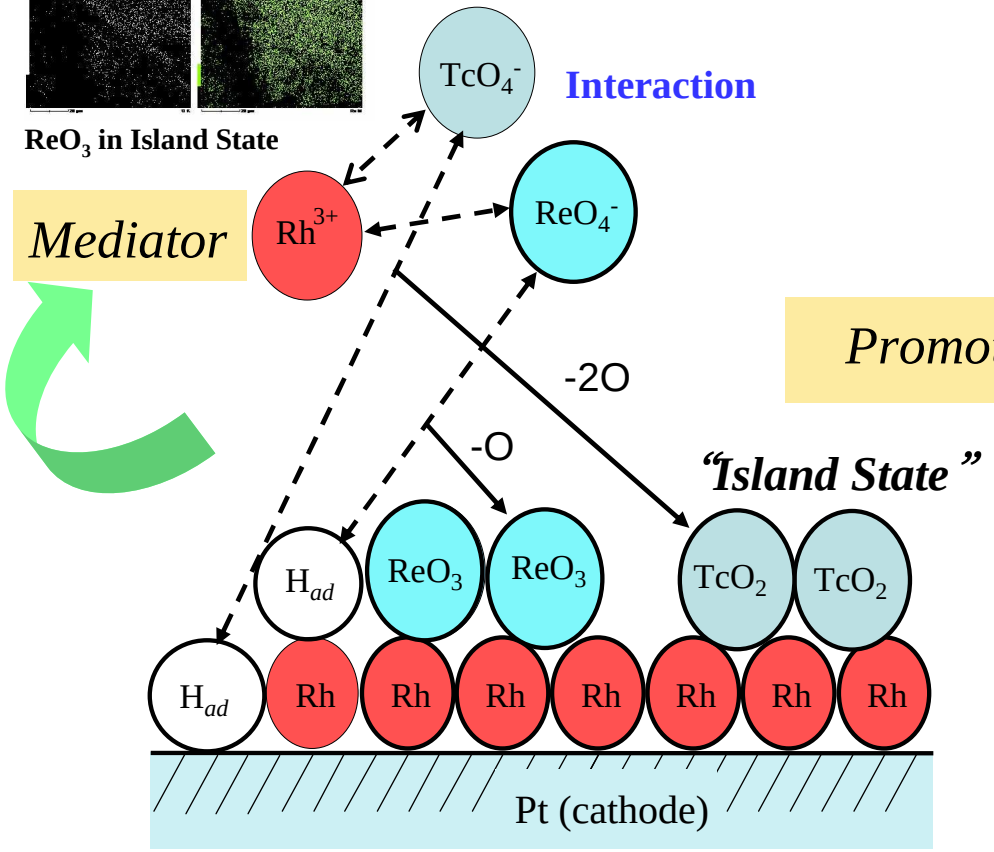
Proposed CEE Model



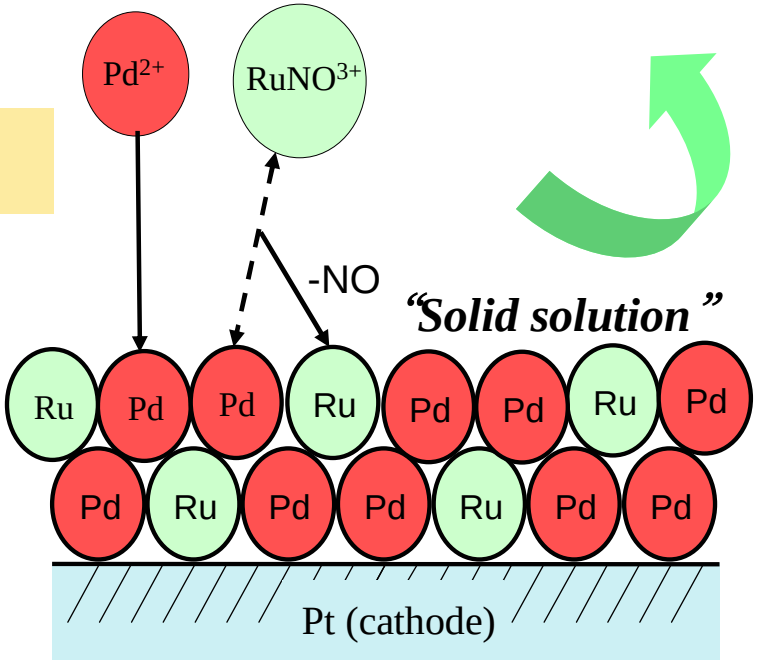
ReO_3 in Island State



Ru/Rh/Pd/Re in Solid Solution

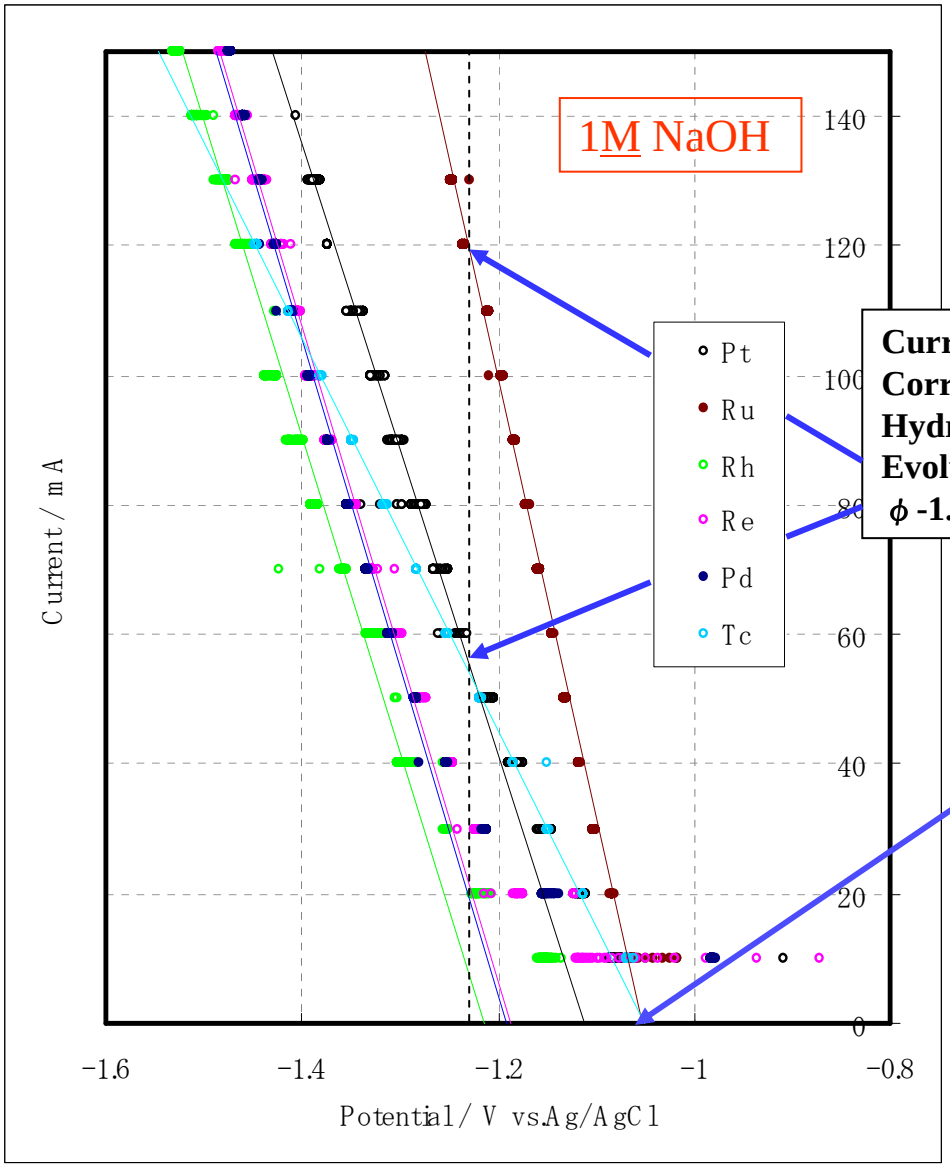


(a) Rh^{3+} as Mediator



(b) Pd_{adatom} as Promoter

Electrochemical Data on RMFP electrodes in 1M NaOH Soln.



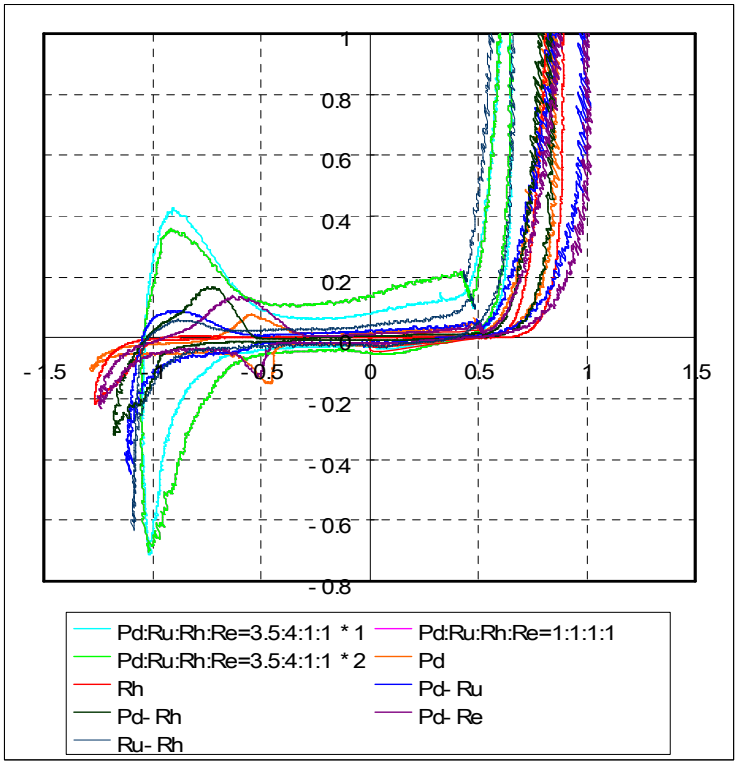
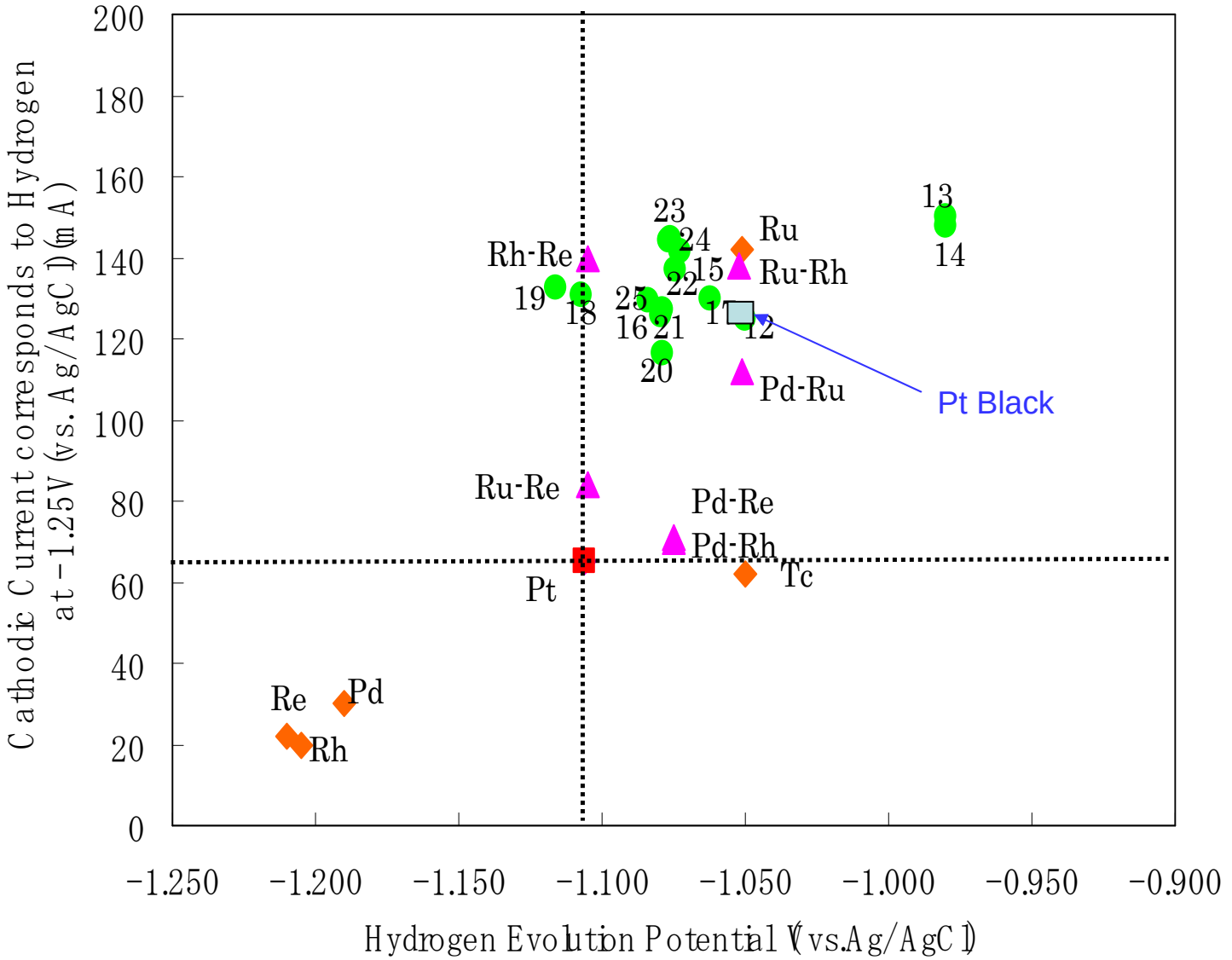


Fig. CV on each deposit electrode in 1M NaOH.

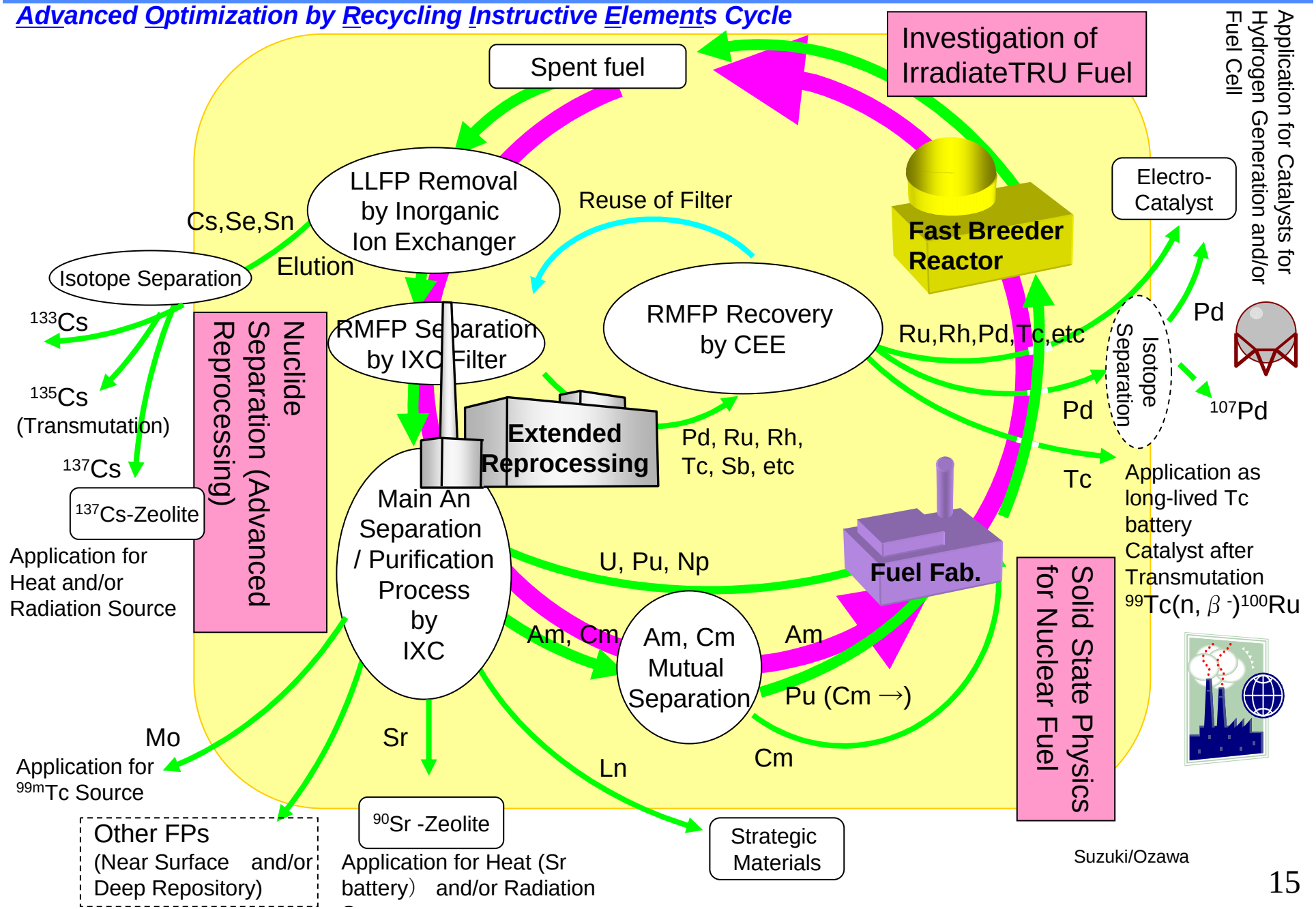
Cathodic Polarization Curves of Pd, Ru, Rh, Re and Tc deposit Pt Electrodes *(left)*, and CV *(right)* *Soln. Composition : 3.5:4:1:1, Pd^{2+} Divided Addition

Electro catalytic Reactivity of RMFP-deposit Pt Electrodes for H_2 Production



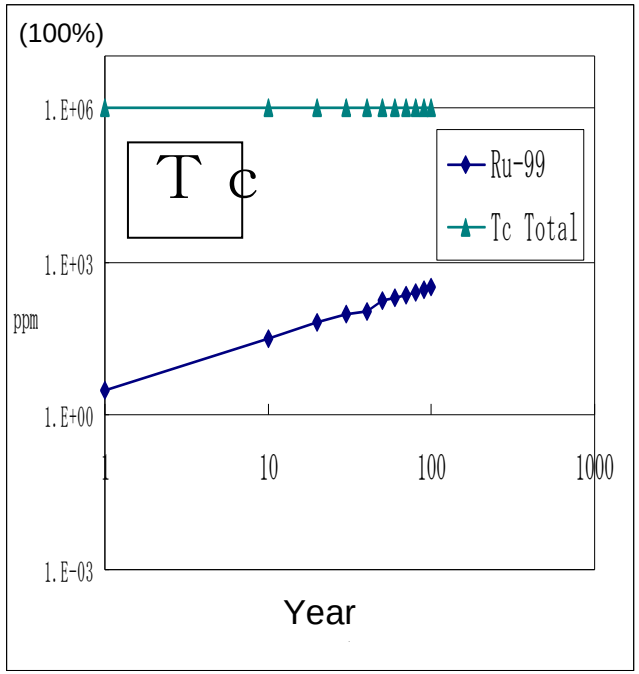
Relation between cathodic current corresponds to hydrogen evolution at -1.25V and initial hydrogen evolution potential ($\phi_{Hinit.}$) on each deposit electrode in 1M NaOH

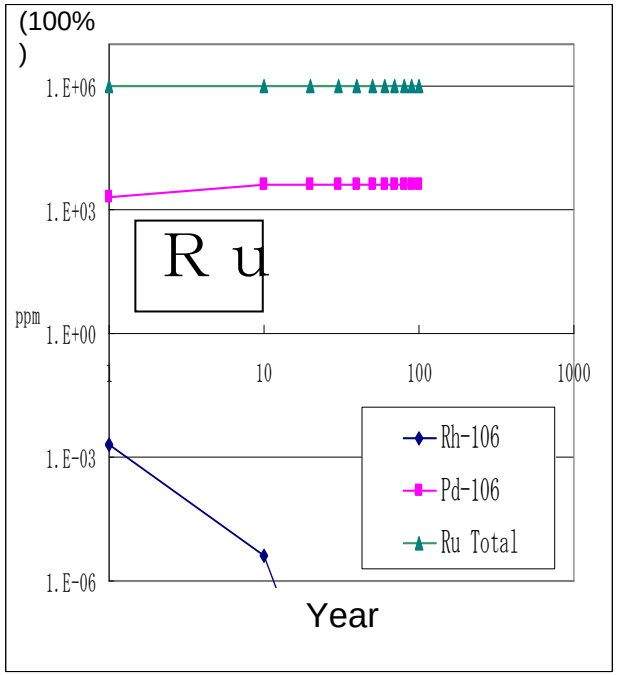
Adv. ORIENT Cycle

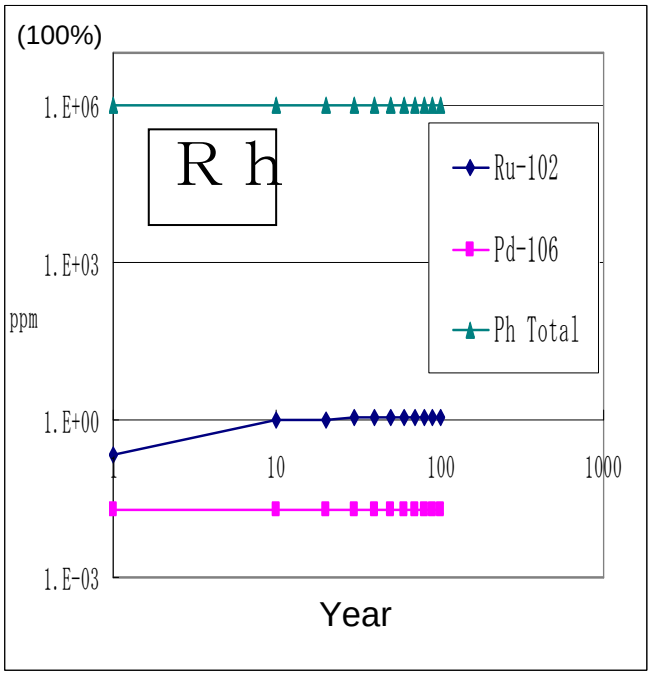


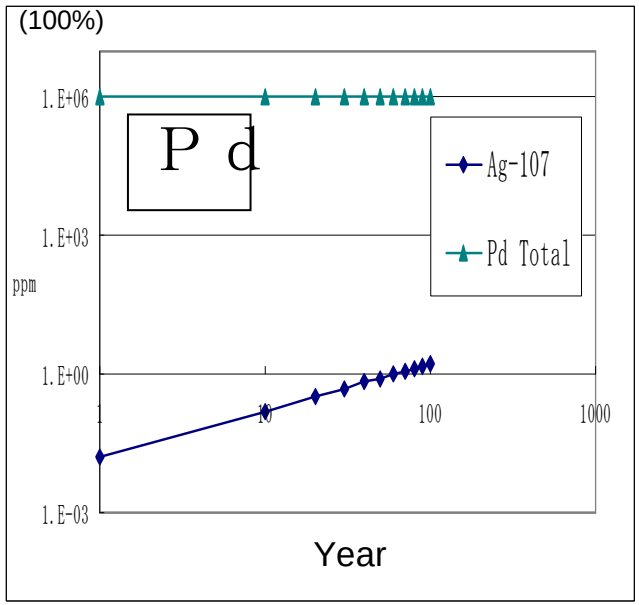
Conclusions

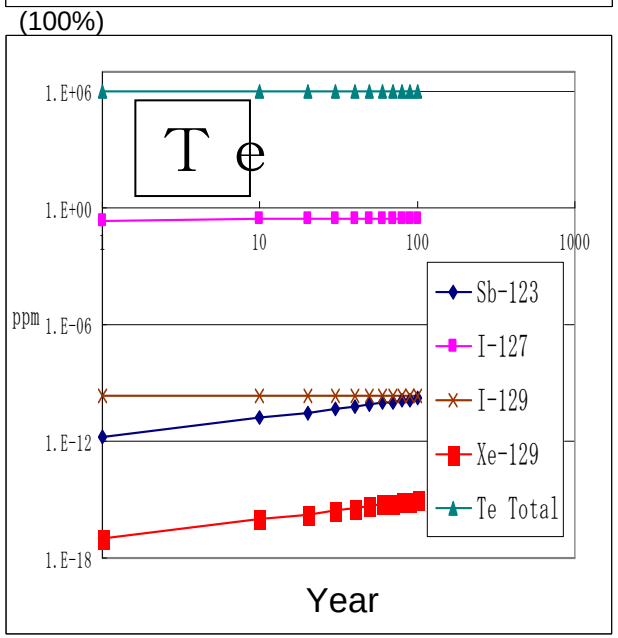
- **Trinitarian research, Adv.ORIENT cycle, program was set forth on separation, transmutation and utilization of instructive materials.**
- **Enhanced element separation of An, LLFP, RMFP and exothermic nuclides is in the duty. Isotope separation of LLFP is also in the scope.**
- **Open up a new forefront field on the chemical / radiochemical utilization of RMFP, Exothermic nuclides, Ln, etc in the industry.**
- **Based on Adv.ORIENT cycle, symbiotic, environmentally-friendly energy system by FBR (Pu) and H_2 can be proposed.**
- **CEE process can recover Ru, Tc and Re as well as Pd and Rh from nitric acid and simulated HLLW. Max. separation ratios of RMFP were, >99% for Pd and Rh, 60% for Ru, 55% for Re and 25% for Tc, respectively. Expecting dramatic increase in hydrochloric acid media.**
- **Quaternary (Ru,Rh,Pd,Re) deposit Pt electrode showed twice as high reactivity vs. smooth Pt electrode on the electrolytic**

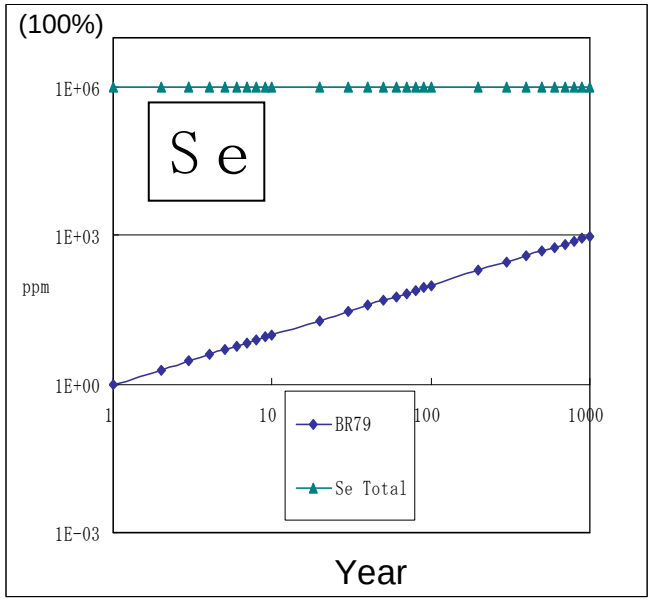

(100%)
Tc
Ru-99
Tc Total
1.E+06
1.E+03
1.E+00
1.E-03
ppm
1
10
100
1000
Year


(100%
)
Ru
Rh-106
Pd-106
Ru Total
1.E+06
1.E+03
1.E+00
1.E-03
1.E-06
ppm
1
10
100
1000
Year


(100%)
Rh
Ru-102
Pd-106
Ph Total
1.E+06
1.E+03
1.E+00
1.E-03
ppm
1
10
100
1000
Year


(100%)
Pd
Ag-107
Pd Total
1.E+06
1.E+03
1.E+00
1.E-03
ppm
1
10
100
1000
Year


(100%)
Te
Sb-123
I-127
I-129
Xe-129
Te Total
1.E+06
1.E+00
1.E-06
1.E-12
1.E-18
ppm
1
10
100
1000
Year


(100%)
Se
BR79
Se Total
1E+06
1E+03
1E+00
1E-03
ppm
1
10
100
1000
Year

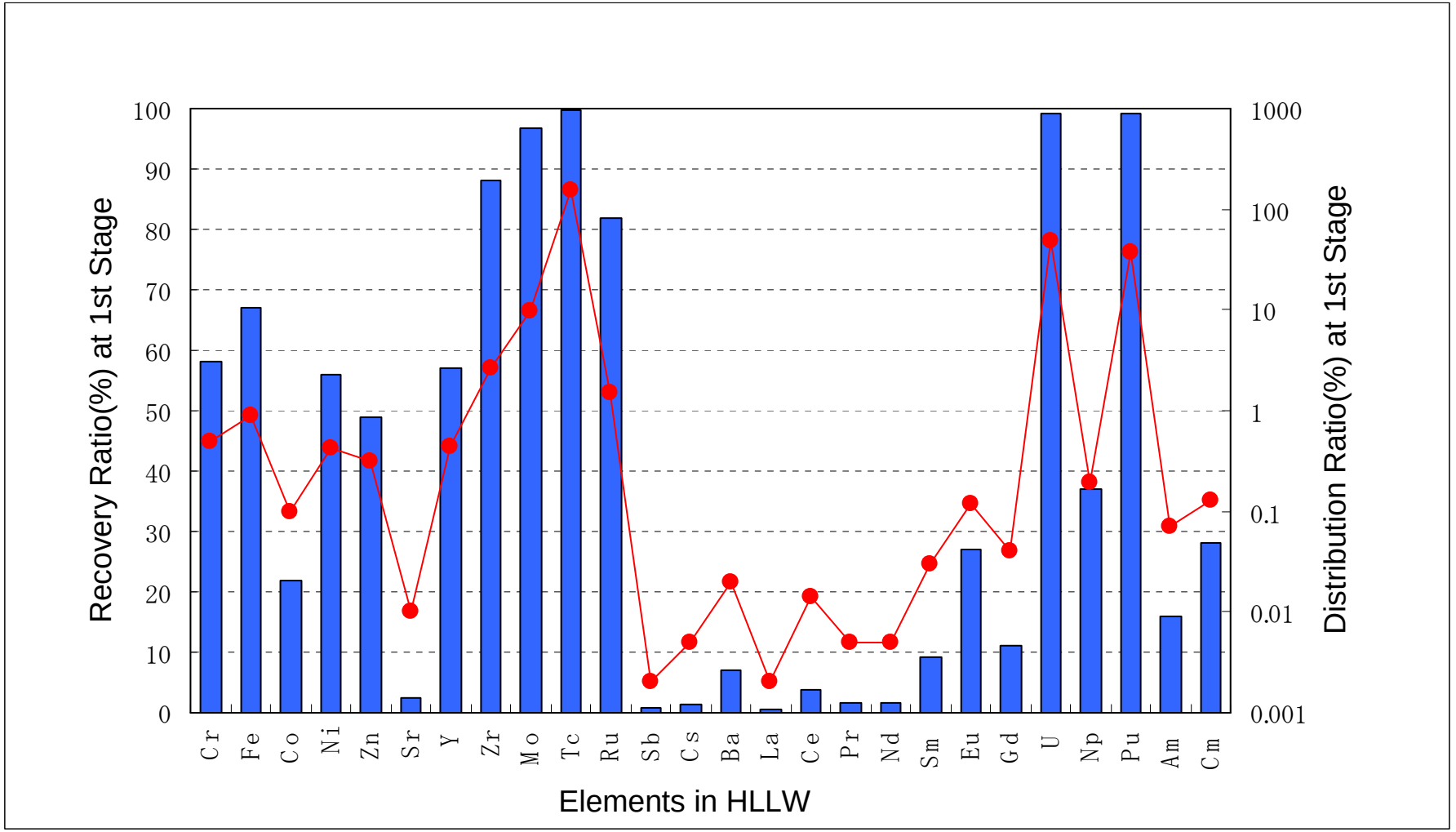


Table 6-8, 6-9 in the KRI Final Report 2005