7th International Symposium on Technetium and Rhenium Science and Utilization

ISTR Retrospectives and Prospects
- In view points of Nuclear Fuel Cycle -

Plenary Lecture

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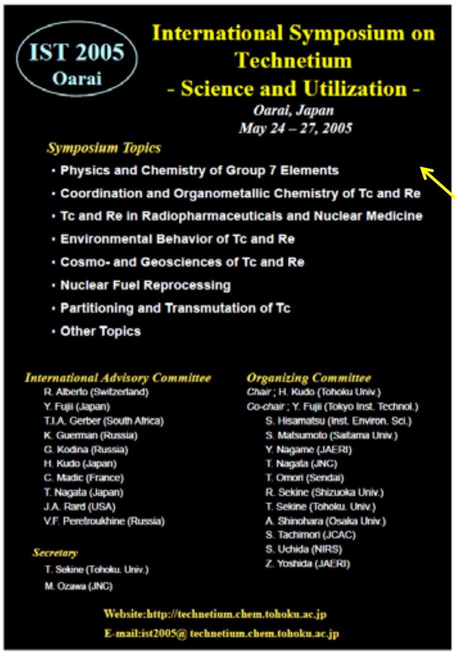
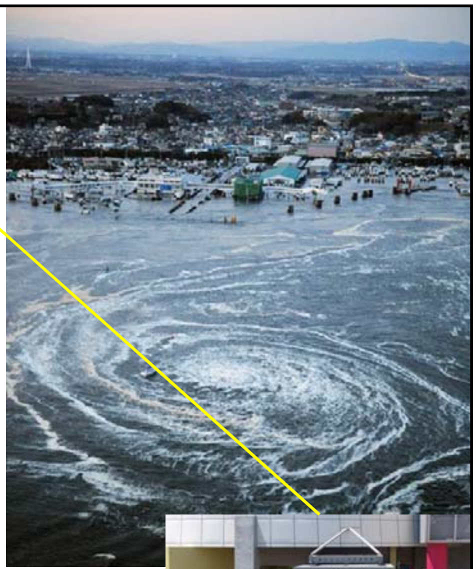
History of Technetium Symposium

1. Tc 1993 – Topical Symposium on the Behavior and Utilization of Technetium (Sendai, Japan, March 18-20, 1993)
2. Tc 1996 – Russian-Japanese Seminar on Technetium (Moscow, Russia, July 1-5, 1996)
3. Tc 1999 – 2nd Japan - Russian Joint Seminar on Technetium (Shizuoka, Japan Nov.29 - Dec.2 1999)
4. Tc 2002 – 3rd Russian - Japanese Seminar on Technetium (Dubna, Russia, June 23 – July 1, 2002)
5. IST 2005 – International Symposium on Technetium – Science and Utilization (Oarai, Ibaraki, Japan, May 24-27, 2005)
6. IST 2008 – 6th International Symposium on Technetium and Rhenium – Science and Utilization (Port Elizabeth, South Africa, October 7-10, 2008) . The 6th delegates honored Prof.Yoshihara for his pioneering role in the Tc chemistry.
7. ISTR 2011 - 7th International Symposium on Technetium and Rhenium – Science and Utilization (Moscow, Russia - July 4-8, 2011)
8. ISTR2014 -8th (USA)

Prof. kenji Yoshihara





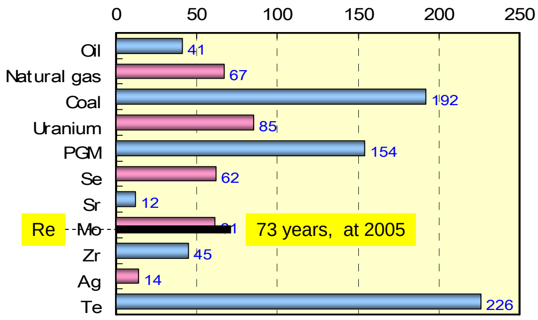
TSUNAMI at O-arai, 2011.3.11

JAEA Chirdren's Science Center, O-arai, was attacked by TSUNAMI. Re-opened on 19th April, 2011

Tc notes

1. Resources
2. Environmental Impact
   - Chernobyl and Fukushima
3. Nuclear Fuel Cycle
   - Reprocessing and Vitrification
4. Utilization
   - Catalyst

Will Tc itself be a Rare Metals, or a substitute for Re ?



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

- Re is the rarest element in the earth and the universe, and will probably be deficient within a 100 years together with Mo.
- On the contrary, Tc is rather rich in the spent nuclear fuel.

•Natural fossil fuels (Oil, Gas) and U are limited in 40-80 years. Rare metals and coal are limited to around 200 years.

•Worldwide CO_2 issue (waste of Oil !) is inevitable.

F/W *Nuclear (FBR) Renaissance*

A/W *Fukushima NPP*

•In turn, PGM seems to be rather abundant, however its resource is extremely localized. Strategy of producing countries will strongly affect the stable supply and prices.

•Rare earth resource is also world-widely localized. About 1% of national GDP will drop if ca.20% of those supply decreased.

F/W *Urban Mine, Nuclear Ore*

Natural Ore and Nuclear Ore

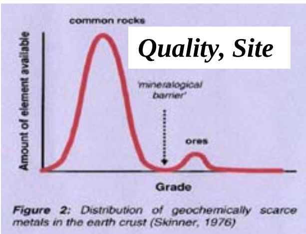


Figure 2: Distribution of geochemically scarce metals in the earth crust (Skinner, 1976)

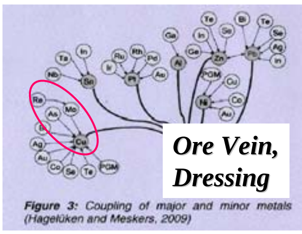
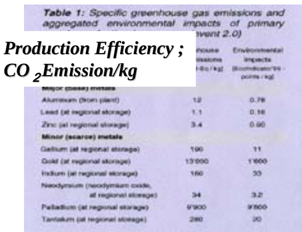


Figure 3: Coupling of major and minor metals (Hagelüken and Meskers, 2009)



P.Wager, et al., Sustainable Governance of Scarce Metals, R'09 Twin World Congress (2009).

Geological, Geopolitical

Technological Factor

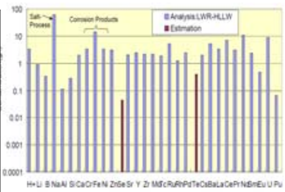
Efficiency & Cost

Spent Fuel

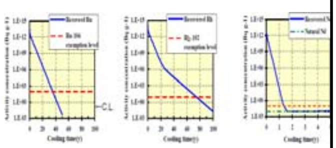
Radiological

LWR-HLLW; ca.10^{10}Bq/ml

Metal	Contents (ppm)	LWR		FBR		Remark
		Conc.(ppm)	Ratio (-)	Conc.(ppm)	Ratio (-)	
Se	(12～92)	50～98	1.4	140	7.1	Estimated from Cu ore in Russia UGMK (2004)
Mo	140	4,021～6,059	36	8,966	84	Results of Erdenet mine in Mongolia
Rh	(0.4～0.6)	578～949	1,527	2,543	6,652	Estimate from PGM production results in main mine
Pd	2.4～7.4	1,900～4,150	617	6,988	1,426	Results of North American palladium Ltd in Canada
Ag	46～201	102～251	1.4	715	5.8	Results of Galmony mine in Ireland and Dikulushi mine in Congo
Te	(3.6～29)	634～842	45	1,840	113	Estimated from Cu ore in Russia UGMK (2004)

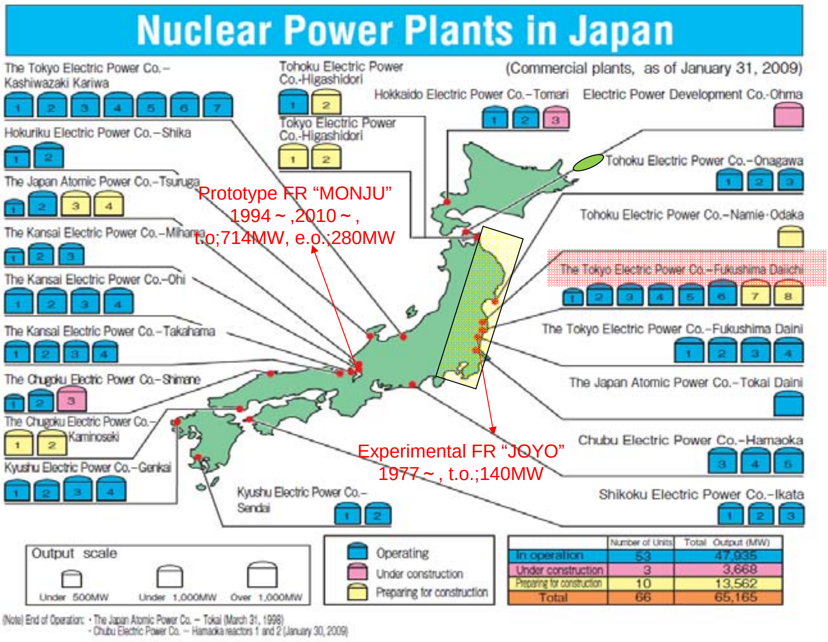


HLLW-LWR (Tokai-Reprocessing Plant)

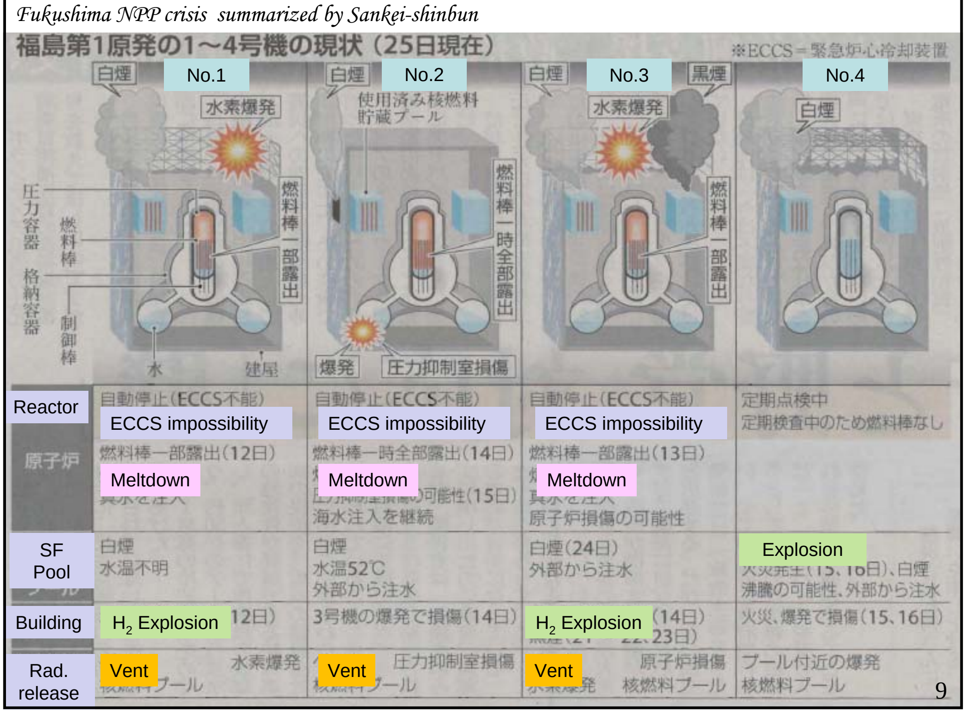


Time Dependency of Bq/g on Nuclear Rare Metals

Fukushima NPP crisis



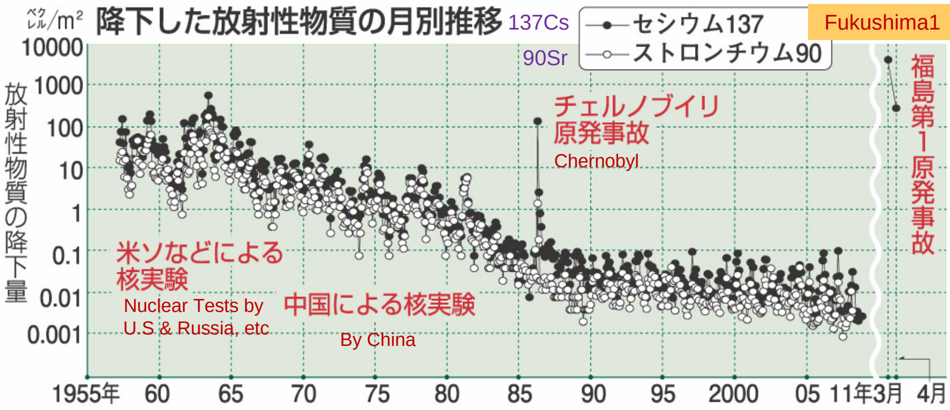
Fukushima NPP crisis summarized by Sankei-shinbun



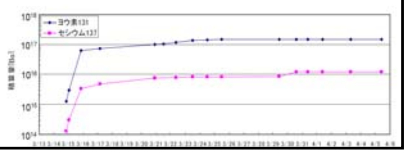
Global Rad. Contamination

Monthly Fallout in Tokyo/Japan Since 1955

Fallout(Bq/m²)



Release of ^{131}I and ^{137}Cs to the atmosphere



Tc released by Chernobyl

Radionuclide released during the Chernobyl Accident

Core inventory on 26 April 1986			Total release during the accident	
Nuclide	Half-life	Activity (PBq)	Percent of inventory	Activity (PBq)
33Xe	5.3 d	6500	100	6500
131I	8.0 d	3200	50 – 60	1760
134Cs	2.0 y	180	20 – 40	54
137Cs	30.0 y	280	20 – 40	85
132Te	78.0 h	2700	25 – 60	1150
89Sr	52.0 d	2300	6	115
90Sr	28.0 y	200	6	10
140Ba	12.8 d	4800	6	240
95Zr	1.4 h	5600	4	196
99Mo	67.0 h	4800	4	168
103Ru	39.6 d	4800	4	168
106Ru	1.0 y	2100	4	73
141Ce	33.0 d	5600	4	196
144Ce	285.0 d	3300	4	116
239Np	2.4 d	27000	4	95
238Pu	86.0 y	1	4	0.035
239Pu	24 400.0 y	0.85	4	0.03
240Pu	6 580.0 y	1.2	4	0.042
241Pu	13.2 y	170	4	6
242Cm	163.0 d	26	4	0.9

total 73559 total 10933 15% released

7.3E19Bq 1.09E19Bq

from Chernobyl: Assessment of Radiological and Health Impact. Chapter 2,
The release,dispersion and deposition of radionuclides,
2002 update of Chernobyl: Ten years on
http://www.oecd-nea.org/rp/chernobyl/c02.html

conversion: 10E15 petabecquerel PBq

γ Contamination of a returned Japanese traveler from Kiev

粒子状の放射性物質が付着 Total 57.1nCi

131I	16 (nCi)	28 (%)
95Zr-95Nb	8	14
103Ru-103mRh	6.5	11.4
141Ce	6.0	10.5
144Ce-144Pr	5.4	9.5
140Ba-140La	5.0	8.8
132Te-132I	2.5	4.4
239Np	2.5	4.4

137Cs-137mBa	0.7	1.2
134Cs	0.4	0.7
99Mo-99mT	0.2	0.4

Tc released by Fukushima

Water

•Analysis of contaminated water at TB of Fukushima 1

Measured 2011/3/24 13 Day after

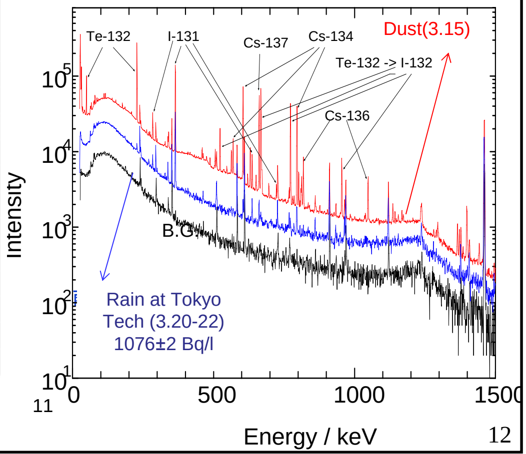
Nuclides	Bq/cc Measured (by TEPCO)	Bq/cc Core Inventory	Percent of Inventory (dissolved ratio)
Y-91			
Co-60	7.00E+02		
Tc-99m	2.50E+03	1.98E+07	0.013%
I-131	1.20E+06	9.93E+07	1.208%
Cs-134	1.80E+06	7.27E+07	2.474%
Cs-136	2.30E+04	1.01E+07	0.227%
Cs-137	1.80E+05	5.88E+07	0.306%
Ba-140	5.20E+04	2.51E+08	0.021%
La-140	9.40E+03	2.89E+08	0.003%
Ce-144	2.20E+06	4.07E+08	0.540%
合計	5.47E+06	1.21E+09	0.453%
I, Cs	3.20E+06	2.41E+08	1.329%

*Tc release from Fukushima NPP 1 is estimated to be very little

Atmosphere

γ- Spectrum of samples at Tokyo

*No ^{241}Am (^{241}Pu) and ^{99}Mo detected at Tokyo



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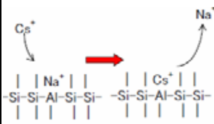
Decontamination of Rad.water in TB

Rad. contaminated Water; ca. 1.1x$10^5$$m^3$

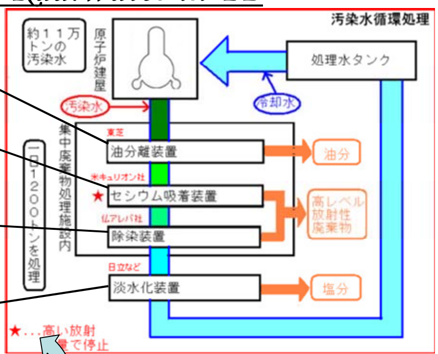


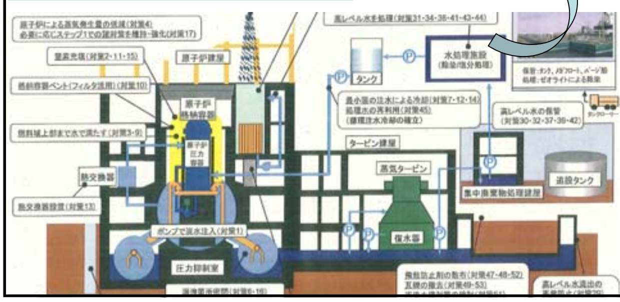
Oil , SS separation

Cs separation
(Ion Exchange Zeolite)

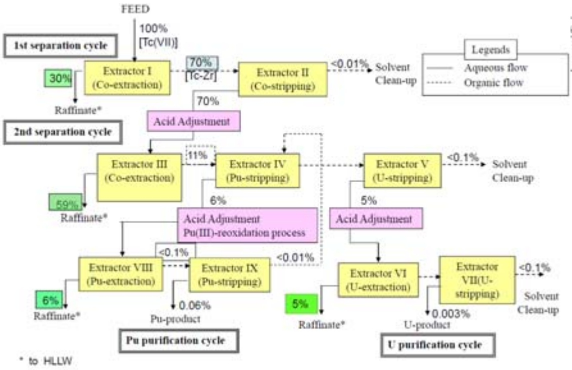
Radioactive separation
(Agglomeration Ni_4 [Fe (CN) $_6$] $_{8/3}$·

Desalting
(Reverse Osmosis Membrane, Evaporation)

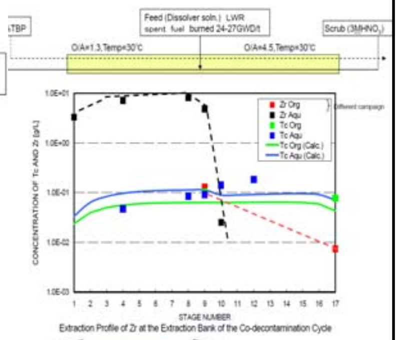




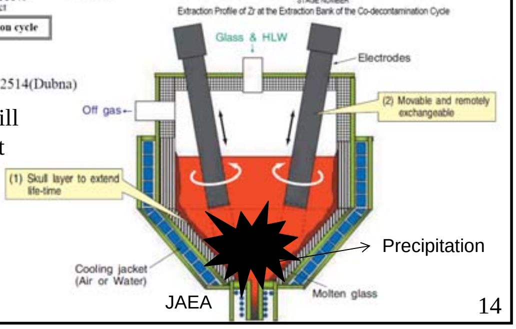
Nuclear Fuel Cycle (Reprocessing, Vitrification)



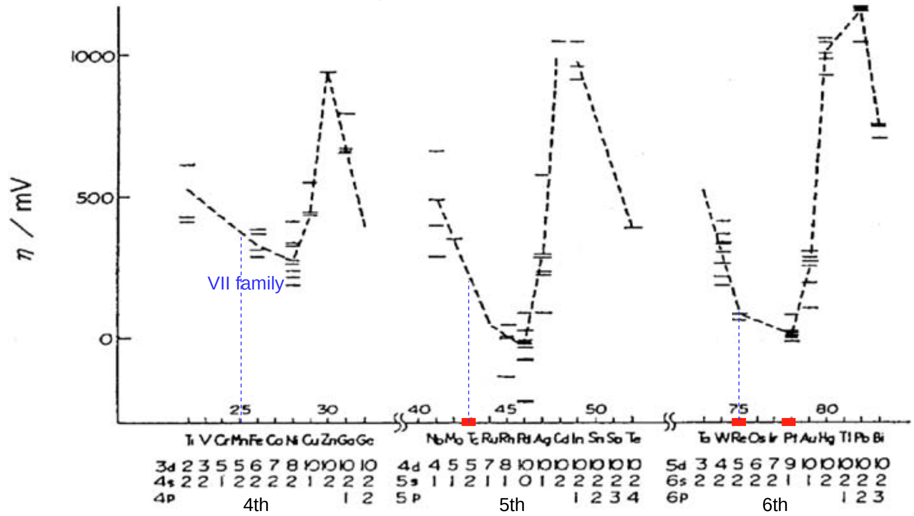
Tc Balance at the Tokai-Reprocessing Plant, Tc2002, Paper No.2514(Dubna)



- PGM elements, especially RuO_2 particle will increase viscosity of melted glass, and might be precipitated at the bottom of melter. RuO_2 also will cause troubles by increasing of electrical conductivity of melted glass.
- Behavior of other PGM , Zr, Mo and TcO_2 must also investigated in this point of view.



Hydrogen Overpotential



Periodicity of Hydrogen Overpotential (Acidic Solution, Ic :1mA/cm²) (Ref.) H.Kita, et al. Denkikagaku, 38,17 (1970)

Tc prospects ,beyond the catastrophe (Conclusion)

1 Separation (Nuclear Fuel Cycle)
- Environmental (land, sea) contamination and remediation, Bioaccumulation
- Reprocessing scheme, especially undissolved residue
- Vitrification behavior; prior separation of PGM, Tc, Mo?

2 Utilization
- Catalyst (Hydrogen production, Fuel-cell ,etc)
- Material (Anti-corrosive, Heat-resistant alloy) , Re substitute
- ^{99m}Tc generation (^{235}Ufission, Mo(n,γ),(n,2n)) for medical use
- β^- source radioisotope micro battery ?, like ^{63}Ni , ^{147}Pm (Cornell Univ.,etc)

3 Transmutation
- ”Raw element” for precious metal production

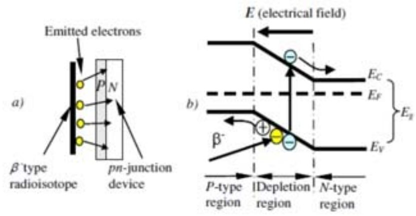
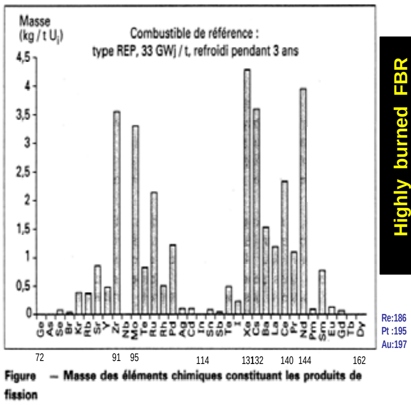


Figure 1. Betavoltaic effect a) Schematic diagram of betavoltaic battery b) Potential diagram of a betavoltaic effect

Nikkei Business Publications

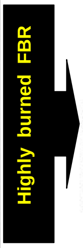
Tc Battery
IPC・RAS

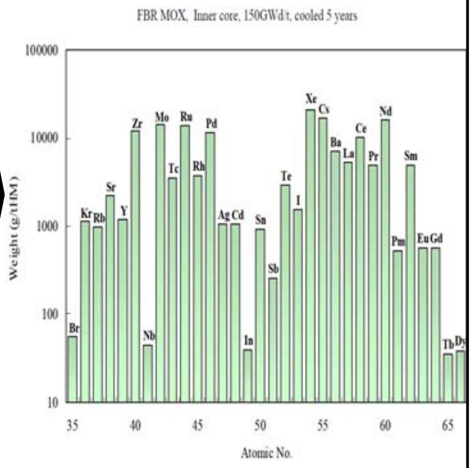
Tc in Nuclear Ore (g/tHM) ; SF-LWR and SF-FBR



Mass of FP elements in irradiated fuel

(M.Bourgeois, Retraitement du combustible, 1994)

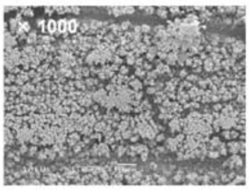




LWR-Reprocessing plant 800 t /y, when recovery ratios will be 80 %,

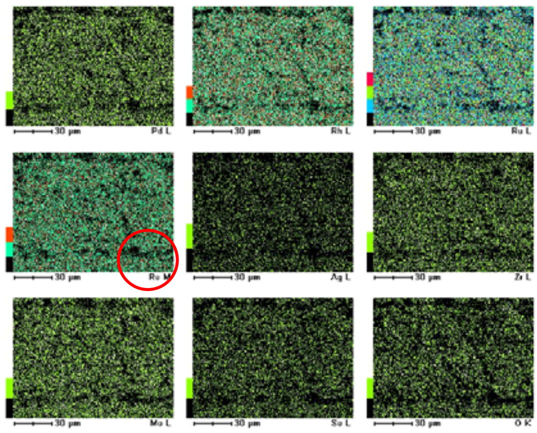
Annual output of NRM will be, Tc: 0.64t, PGM: 2.56t, Lns: 6.4t, Mo:1.92t, etc

Micro NRM Deposits by CEE of S-HLLW$_{HCl\text{-}0.5M}$



Mixture of Dendrite and Coagulated fine sphere particles

SEM/EDS Analysis



- PGM,Tc, Re co-deposit electrodes showed higher catalytic activity than Pt-black on H_2 production.
- Tc showed the same or higher reactivity than that of Re in these co-deposit systems.

■ Metal ; Ru,Pd,Rh, Oxide ; ReO_3, TcO_2, MoO_2
■ Ru, Rh, Re > Pd, Mo