極低バックグラウンド素粒子原子核研究懇談会 @富山 Apr 23, 2013

Enrichment Review ~Enrichment of ^{48}Ca" ~

D_2O 0.015 % → 99.92% Distillation/Electrolysis, etc. C$330M

Thanks to T. Sato, T. Oi @Sophia Univ. & M. Nomura @TIT, Japan
Do-Young Jeong & Cheol-Jung Kim@KAERI, Korea
H. Niki @ Univ. of Fukui, Japan
Elena A. Nikitina, Kolygaeva Galina@ECP, Russia
Chang-Hong Yu @ORNL, USA
Steve Elliott @LANL, USA
Giorgio Gratta @Stanford, USA

R. Hazama
Osaka Sangyo University

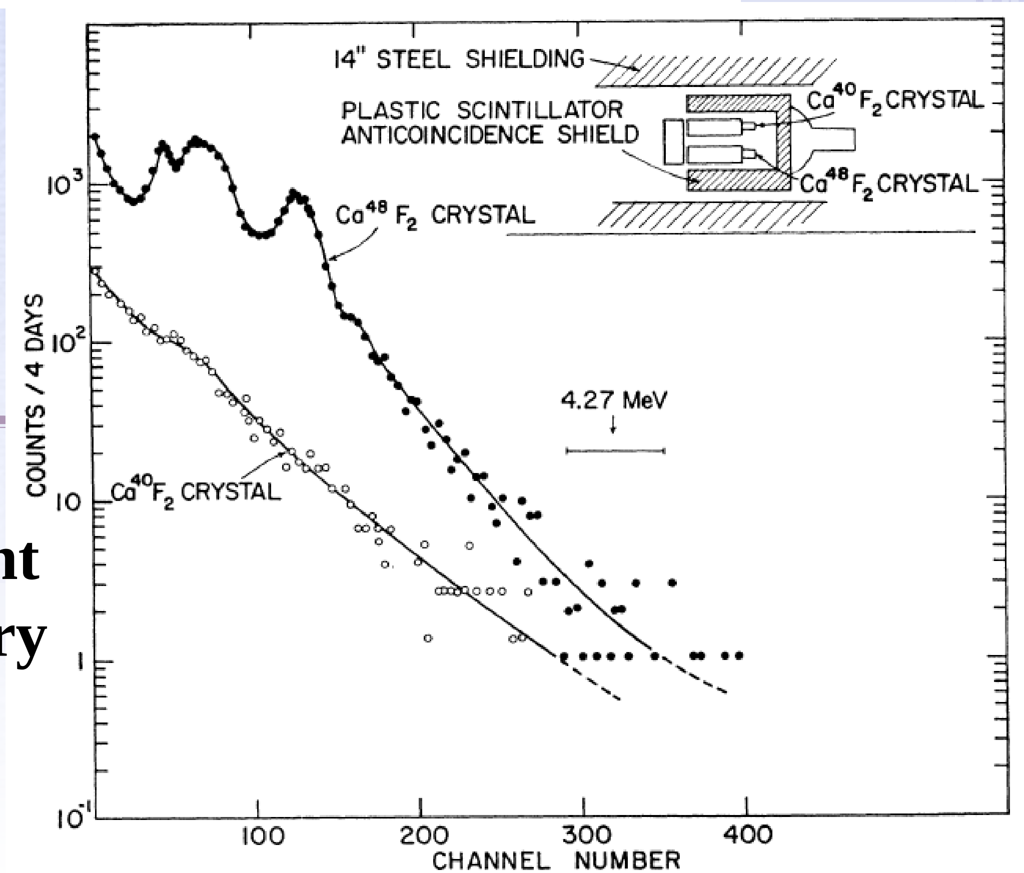
First experimental results

Limits for Lepton-Conserving and Lepton-Nonconserving Double Beta Decay in Ca^{48}†

E. DER MATEOSIAN AND M. GOLDHABER
Brookhaven National Laboratory, Upton, New York
(Received 10 February 1966)

PHYSICAL REVIEW 146 (1966) 810

$^{48}CaF_2(Eu)$, 11.4 g
$\rightarrow T_{1/2}^{0\nu} > 2\times10^{20}$ yr



96.59 % enriched ^{48}Ca
Isotope Separation Department
Oak Ridge National Laboratory

Enriched scintillators for 2β decay search

Year	Scintillator	Ref.
1966	$^{48}CaF_2(Eu)$ (δ=97%, m=22 g) $^{40}CaF_2(Eu)$ (δ=97%, m=22 g)	[1]
1987 – 2003	$^{116}CdWO_4$ (δ=83%, m=510 g)	[2]
2009 – present	$^{106}CdWO_4$ (δ=66%, m=231 g)	[3]
2010 – present	$^{116}CdWO_4$ (δ=82%, m=1868 g)	[4]
2010 – present	$^{40}Ca^{100}MoO_4$ (δ=96% of ^{100}Mo, δ=99.964% of ^{40}Ca depleted on ^{48}Ca, m=550 g)	[5]
2013 – 2014	$Zn^{100}MoO_4$, $Zn^{82}Se$	[6,7]

[1] E. der Mateosian, M. Goldhaber, Phys. Rev. 146 (1966) 810.
[2] F.A. Danevich et al., JETP Lett. 49 (1989) 476; Phys. Rev. C 68 (2003) 035501.
[3] P. Belli et al., Nucl. Instr. Meth. A 615 (2010) 301; Phys. Rev. C 85 (2012) 044610.
[4] A.S. Barabash et al., J. Instrum. 6 (2011) P08011.
[5] S.J. Lee et al., Astropart. Phys. 34 (2011) 732; AMoRE project.
[6] J.W. Beeman et al., Phys. Lett. B 710 (2012) 318.
[7] F. Ferroni, Il Nuovo Cim. C 5 (2010) 27; LUCIFER project.

The industrial separator SU20
Lesnoy, Sverdlovky region

30 kg of Ca-40 ($^{40}CaCO_3$) is available now

Ca-48 < 0,001%

Good for 150 kg of $^{40}Ca^{100}MoO_4$

Productivity: 4 – 5 kg/year

Electromagnet of the SU20 separator

(5-floors building & 3000 ton magnet!)

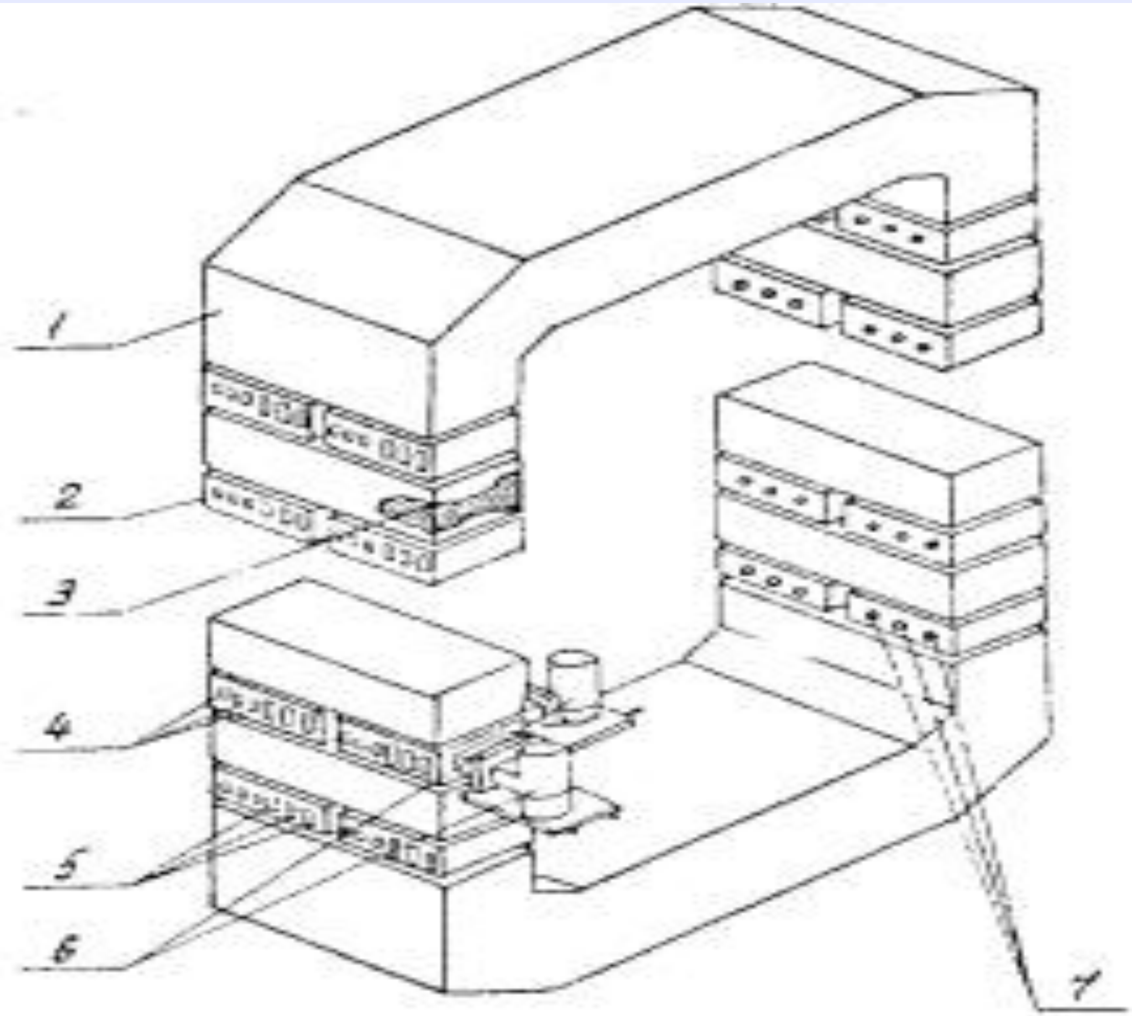


Fig. 1. Electromagnet of the separator. (1) Housing; (2) separation tank (chamber); (3) electromagnet coils; (4) ion source seats; (5) ion receiver seats; (6) diffusion pumps; (7) observation openings.

V.N.Kornoukhov

SU20: Separation tank (2*2*5 = 20)

R = 90 cm

U = 30 kV

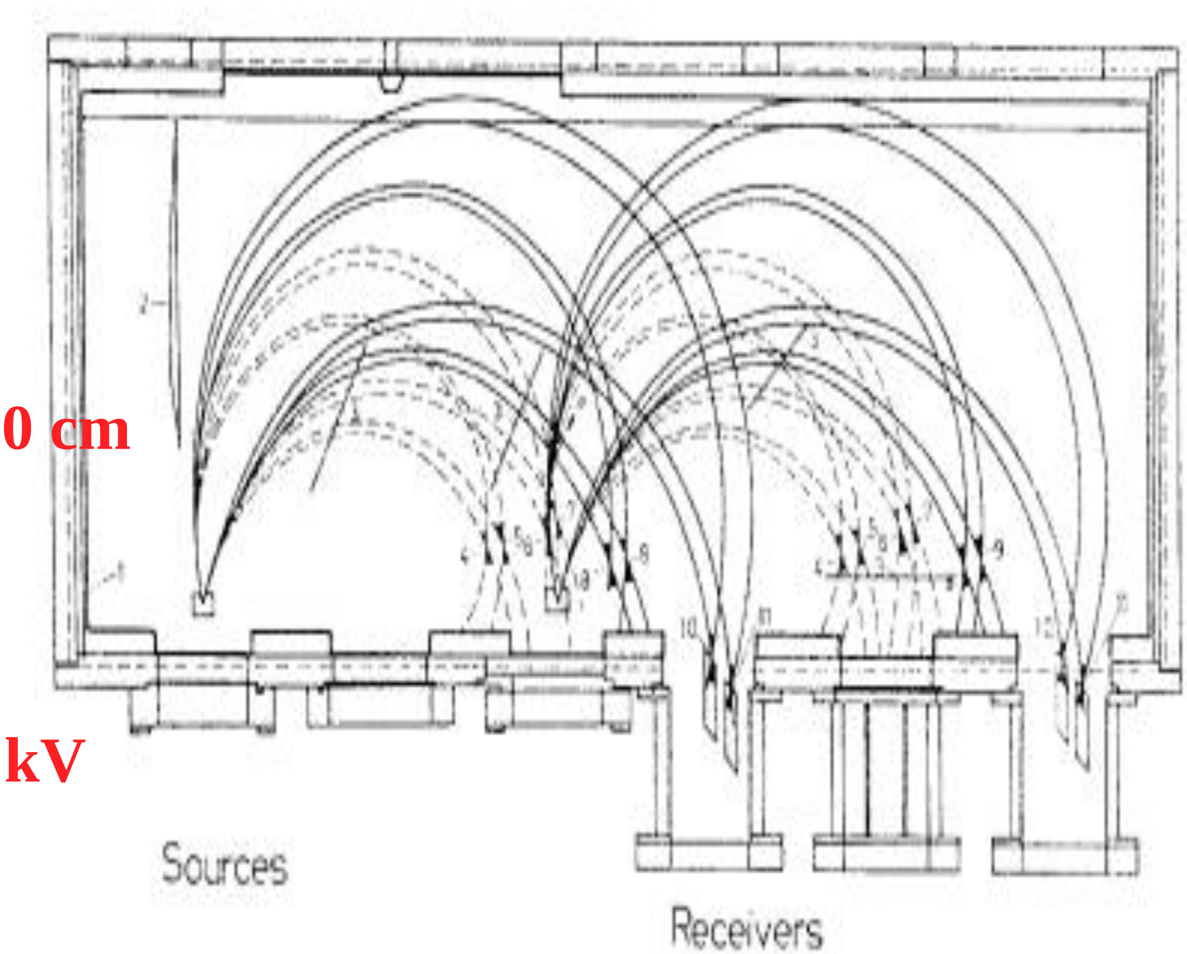


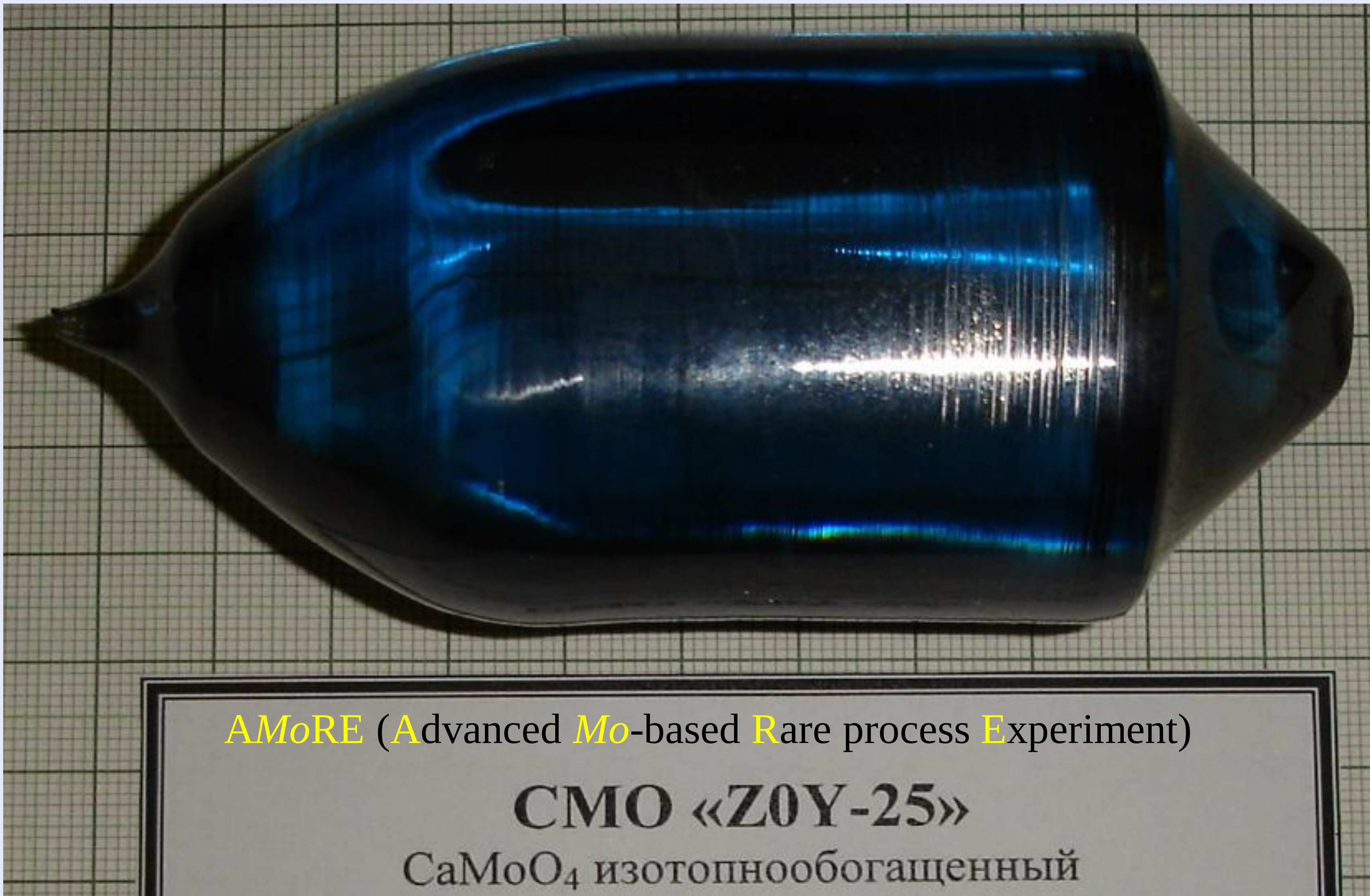
Fig. 3. Location of ion beams in a separation tank, two ion sources operating (feed material is $FeCl_3$): (1) liner; (2) magnet plate profile; (3) graphite plates; (4)–(11) isotope beams: [(4) $^{54}Fe^{3+}$, (5) $^{58}Fe^{3+}$, (6) $^{54}Fe^{2+}$, (7) $^{58}Fe^{2+}$, (8) $^{35}Cl^{+}$, (9) $^{37}Cl^{+}$, (10) $^{54}Fe^{+}$, (11) $^{58}Fe^{+}$].

V.N.Kornoukhov

$^{40}Ca^{100}MoO_4$ single crystal before annealing

(m = 0,55 kg, D49 x 42 mm, $L_{cylinder}$ = 53 mm grown 11/09/2009)

V.N.Kornoukhov



AMoRE (Advanced *Mo*-based Rare process Experiment)

Past Neutrino-less double beta decay searches

Nucleus	Experiment	%	$Q_{\beta\beta}$	Enr	Technique	$T_{0\nu}$ (y)	$<m_{\nu})$
^{48}Ca	Elegant IV	0.19	4271		scintillator	$>1.4x10^{22}$	7-45
^{76}Ge	Heidelberg-Moscow	7.8	2039	87	ionization	$>1.9x10^{25}$	.12 - 1
^{76}Ge	IGEX	7.8	2039	87	Ionization	$>1.6x10^{25}$	.14 – 1.2
^{76}Ge	Klapdor et al	7.8	2039	87	ionization	$1.2x10^{25}$	.44
^{82}Se	NEMO 3	9.2	2995	97	tracking	$>1.x10^{23}$	1.8-4.9
^{100}Mo	NEMO 3	9.6	3034	95-99	tracking	$>4.6x10^{23}$	.7-2.8
^{116}Cd	Solotvina	7.5	3034	83	scintillator	$>1.7x10^{23}$	1.7 - ?
^{128}Te	Bernatovitz	34	2529		geochem	$>7.7 \times 10^{24}$	.1-4
^{130}Te	Cuoricino	33.8	2529		bolometric	$>2.4x10^{24}$	.2-1.
^{136}Xe	DAMA	8.9	2476	69	scintillator	$>1.2x10^{24}$	1.1 -2.9
^{150}Nd	Irvine	5.6	3367	91	tracking	$>1.2x10^{21}$	3 - ?

10.96 kg ←

~1 kg

~7 kg

6.5 kg LXe

NEMO3
36.55 g

The crown jewels of EXO

200 kg of xenon enriched to 80% in ^{136}Xe:
the most isotope in possession by any ββ0ν collaboration.

Giorgio Gratta

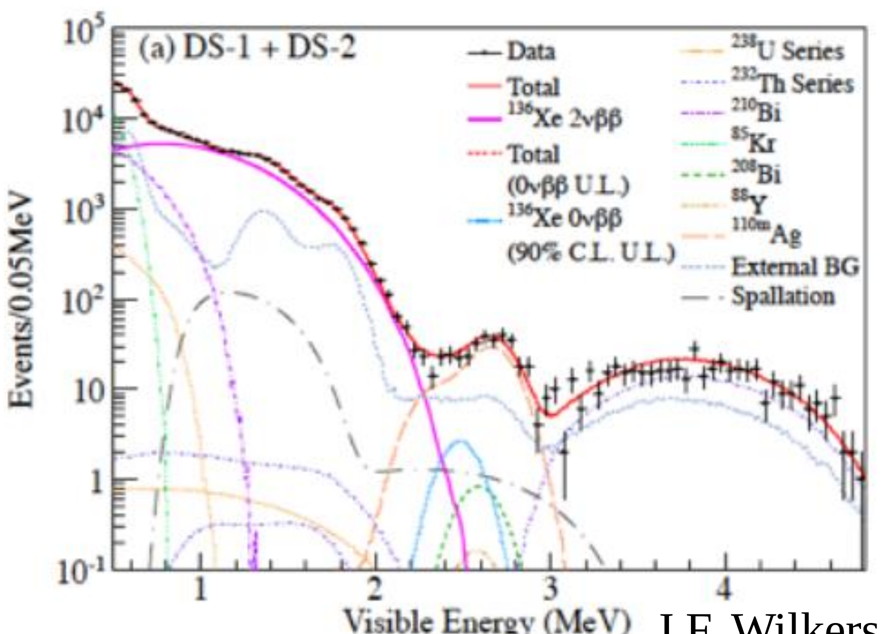
KamLAND-ZEN ^{136}Xe (Japan - U.S.)

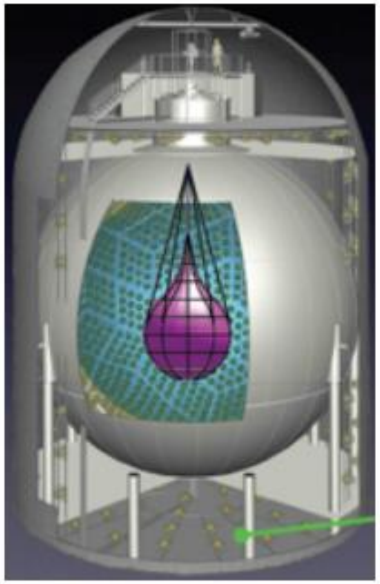
- Utilizes substantial investment and expertise in KamLAND
- ~~300~~ kg of 91.7% ^{136}Xe (2.7% by wt. in liquid scintillator), ~~400~~ kg in hand.
- mini-ballon of R=1.7m, 25 microns thick.
- Since recent result, with 110Ag, working on purification
- Nov. 2012 - fire in purification system area.
- Rebuilding plan is still unclear mostly due to an administrative procedure for the contract.
- Earliest possibility was to restart purification in May, but likely there will be an additional two month delay.
- Longer term plan upgrade to 700 kg

320 kg

620 kg

将来
800 ~ 1 ton





J.F. Wilkerson, NuMass 2013@Milano, Feb 2013

B.A. Barabash, J. Phys. G: Nucl. Part. Phys. 39(2012)085103

Isotope enrichment Price Level

Isotope	Criteria for the Best 0νββ Isotope						0νββ Project
	$Q_{\beta\beta}$ (MeV)	$G_{0\nu}$ (y^{-1})	$T^{2\nu\beta\beta}_{1/2}$ (10^{20} y)	Isotope Enrichment			
				Abundance (%)	Method	Price Level	
^{130}Te	2.533	1.70	6.8	33.8 → 95	GC	0.3	CUORE
^{136}Xe	2.462	1.81		8.9 → 90	GC	0.2	EXO
^{76}Ge	2.039	0.24	15	7.8 → 90	GC	1 ($80/g)	GERDA MAJORANA
^{82}Se	2.995	1.08	0.92	9.2 → 90	GC	1.5	SuperNEMO
^{100}Mo	3.034	1.75	0.07	9.6 → 90	GC	1	AMORE
^{116}Cd	2.802	1.89	0.28	7.5 → 90	GC	2.5	
^{48}Ca	4.274	2.44	0.44	0.187 → 25	EMIS ALSIS	160 < 5	CANDLES
^{150}Nd	3.667	8.00	0.08	5.6 → 90	EMIS	170	
^{96}Zr	3.350	2.24	0.23	2.8 → 60	EMIS	400	-

Do-Young Jeong, KAERI

Method of isotopes separation

- Electro-magnetic method
- Physical Chemical (rectification, chemical exchange)
- Gas centrifuges
- Optical methods (AVLIS) (MLIS) Photochemistry
- Plasma ICR method

H							He		
Li	Be	B	C	N	O	F	Ne		
Na	Mg	Al	Si	P	S	Cl	Ar		
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni
Cu	Zn	Ga	Ge	As	Se	Br	Kr		
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd
Ag	Cd	In	Sn	Sb	Te	I	Xe		
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt
Au	Hg	Tl	Pb	Bi	Po	At	Rn		
Fr	Ra	Ac	Rf	Db	Sg	Bh	Hs	Mt	110
111	112	(113)	114	(115)	116	(117)	118		

La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

Actual production capability

USA:

Calutron production was stopped in 2004

Medium size ICR machine founded by DOE is installed at Theragenics

production is oriented to medicine application

New program for AVLIS founded at Livermore (very expensive)

it is unclear if this program will be completed

Russia:

Few labs are able to produce isotopes with Ultracentrifuges

Only elements that have gas compounds can be produced

Prices of enriched isotopes are favorable (today)

Europe and Japan:

There are some enrichment facilities based on Ultracentrifuges

Restart of an AVLIS machine in France is not yet established (^{150}Nd)

Actually practically only Russian labs can produce stable isotopes

ECP, Zelenogorsk

Two facilities have shown interest in providing DBD isotopes

- ECP of Zelenogorsk supplied all the isotope for IGEX and HM ^{76}Ge DBD experiments
- ECP has quoted Ge prices to Majorana and recently sold Ge to GERDA
  - Quote 2001:
    - $55/g at low volumes
  - Sale to GERDA 2004:
    - $51/g for ~44kg (~2M$)
  - Sale to Majorana 2011:
    - $90/g at 42.5 kg (86%)
- ECP also developed shipping and storage solutions

- Angarsk Electrolytic Chemical Combine
  - (AEKhK) in Irkutsk, http://www.aecc.ru
- ✓ Urals Electrochemical Combine (Sverdlovsk-44)
  - (UEKhK) in Novouralsk, http://www.ricon.e-burg.ru
- Siberian Chemical Combine (Tomsk-7)
  - (SKhK) in Seversk, http://www.atomsib.ru
- ✓ Electrochemical Plant or ECP (Krasnoyarsk-45)
  - (EKhZ) in Zelenogorsk http://www.ecp.ru

UEKhK, Urals

Chang-Hong Yu @ ORNL

ECP PARTICIPATION IN SCIENTIFIC RESEARCH EXPERIMENTS

Since the early 90s the ECP is the supplier of isotopes used under several programs of material structure physical research.

Collaboration (project)	Program content	Year	Isotope	Enrichment	Quantity
IGEX (USA, Russia)	Neutrinoless double beta decay study	1990 – 1992	Ge-76	> 86%	20 kg
NEMO (France, Russia)	Neutrinoless double beta decay study	1990 – 1994 1995	Mo-100 Se-82	> 95% > 95%	13 kg 1 kg
XMASS (Japan)	Solar neutrino registration	2001	Xe – 129 + Xe – 131	> 80%	10 kg
EXO (USA, Russia)	Neutrinoless double beta decay study	2002 – 2003	Xe – 136	> 80%	68 kg
GERDA (Germany, Russia)	Neutrinoless double beta decay study	2004 – 2005	Ge – 76	> 86%	30 – 40 kg
XENON (USA, Italy)	Neutrinoless double beta decay study	2007	Xenon with displaced isotopic composition		5000 L
RAS INR, RAS IMTPM ACTC (Russia)	Neutrinoless double beta decay study	2008	Mo – 100	> 95%	2,5 kg
ITEP (Russia)	Neutrinoless double beta decay study	2008	Sn – 112	> 94%	50 g
Baksan Neutrino Observatory INR RAS (Russia)	Search for Kr-78 2K-capture	2002	Purification of Kr-78>99.8% from Kr-85 – Specific activity of Kr-85 in Kr-78 has been reduced by more than 100 times and is now less than 0.1 Bq/l [5].		
WARP (Italy, Russia)	Dark matter search experiment	2006 – 2007	Purification of argon from Ar-39 is improved by more than 50 times, and from Kr-85 by no less than 100 times as compared with the natural content		
MAJORANA (USA, Russia)	Neutrinoless double beta decay study	2010 2011 July 2012 – plan	Ge – 76	> 86%	10 kg 20 kg 20 kg – plan
MAJORANA (USA, Russia)	Neutrinoless double beta decay study	2010	Te – 130	92 %	10 kg

Year	Is…	…ration …)
1990-1992	G	IGEX Spain,Russia, …rmenia)
1990s	M	KAERI …uth Korea)
1990-1994	M	NEMO …nce, Russia)
1995	S	
2001	X + X	XMASS (Japan)
2002-2003	X	EXO …A, Russia)
2001, 2003	S S S	…nce Berkley …al Laboratory (USA)
2004-2005	G	GERDA …any, Russia)
2002	K 9 K 1 th	…an Neutrino …servatory …NR RAS (Russia)

for Ca

Technologies for isotope production

Find a cost-effective & efficient way of enrichment!!!

Separation technology	*Field of use*	*Production per year*	*Cost*
Electromagnetic (mass-spectroscopy effect)	universal	tens of grams 0.001	high
Chemical & phys. processes (rectification, chem. exchange etc)	light elements	tons	low
Gas diffusion	elements forming gas compounds	thousands of tons	middle
Gas centrifuge	elements forming gas compounds	thousands of tons 1	low
Laser (optical) separation	elements having isotope shift of spectrum lines	kilograms 0.1	middle
Plasma ion-cyclotron effect (under developing – the USA, Russia)	universal	hundreds of kilograms 0.01	middle

- Liquid centrifuge? (mobility/viscosity with $CaCl_2$ solution & almina)
- Gel electrophoresis ($CaCl_2$ & HCl)
- Electro-migration

B.A. Barabash, J. Phys. G: Nucl. Part. Phys. 39(2012)085103

Crown Ether

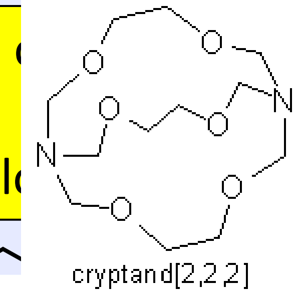
Liquid

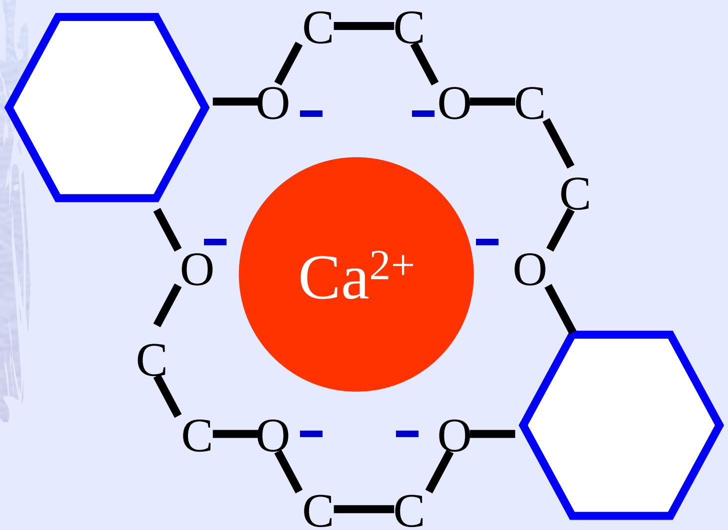
Microchip

Soild

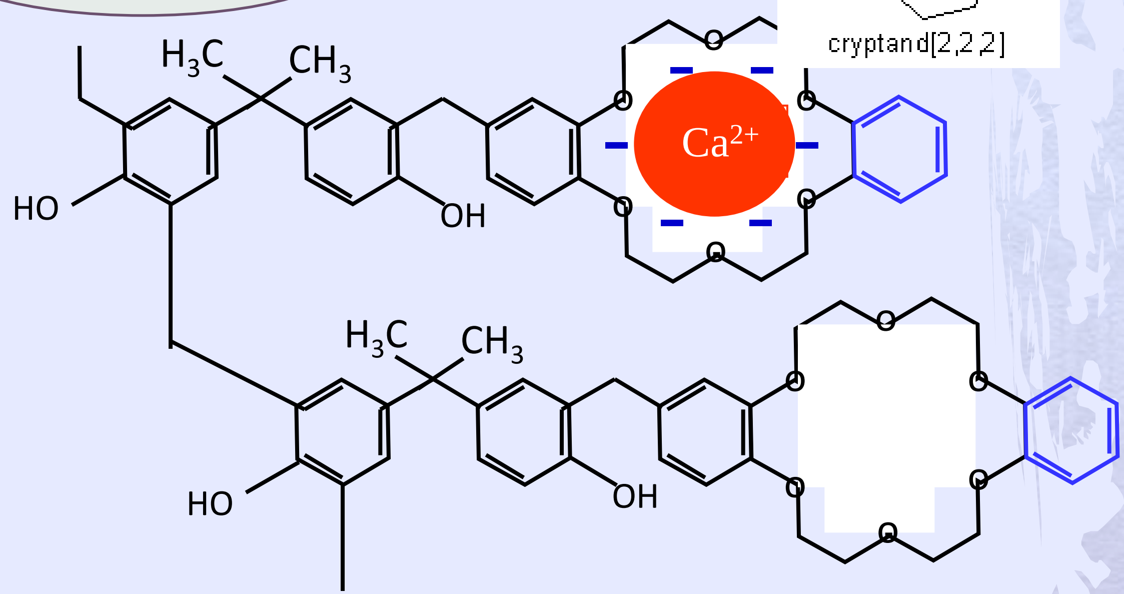
Resin

Benzo
or
Di-cycl ... C6





Dicyclohexano 18-crown-6

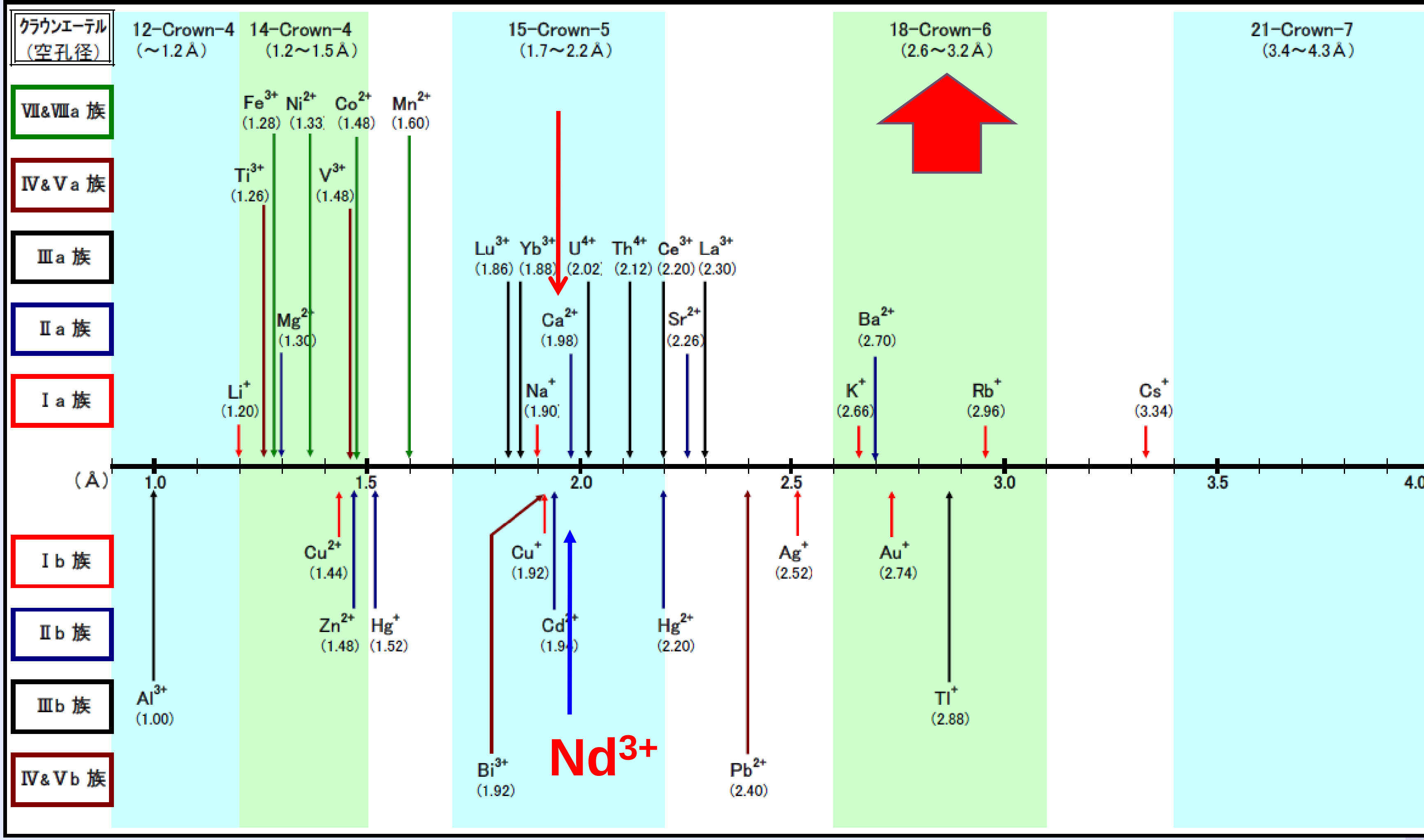


DC18C6

Total # of atoms in the ring

# of oxygen atoms in the ring

- **Held by electrostatic attraction between negatively charged O^- of the C-O dipoles & cation (Ca^{2+})**
- **How well the cation fits into the crown ring**
- **Liquid(aq-salt)-liquid(org-crown)/solid(resin) extraction in isotopic equilibrium**



Ca isotope	^{40}Ca	^{42}Ca	^{43}Ca	^{44}Ca	^{46}Ca	^{48}Ca
abundance（%）	96.94	0.65	0.135	2.09	0.004	0.187

Solid./Liq.

Increase Ca content → ε doesn't change

A-type FH-type

	)	Run (3)	
Con in the		0.25	0.088
	6	0.00211	0.0034
	45	0.00026375	0.000425
lo	2	−3.5788	-3.3716

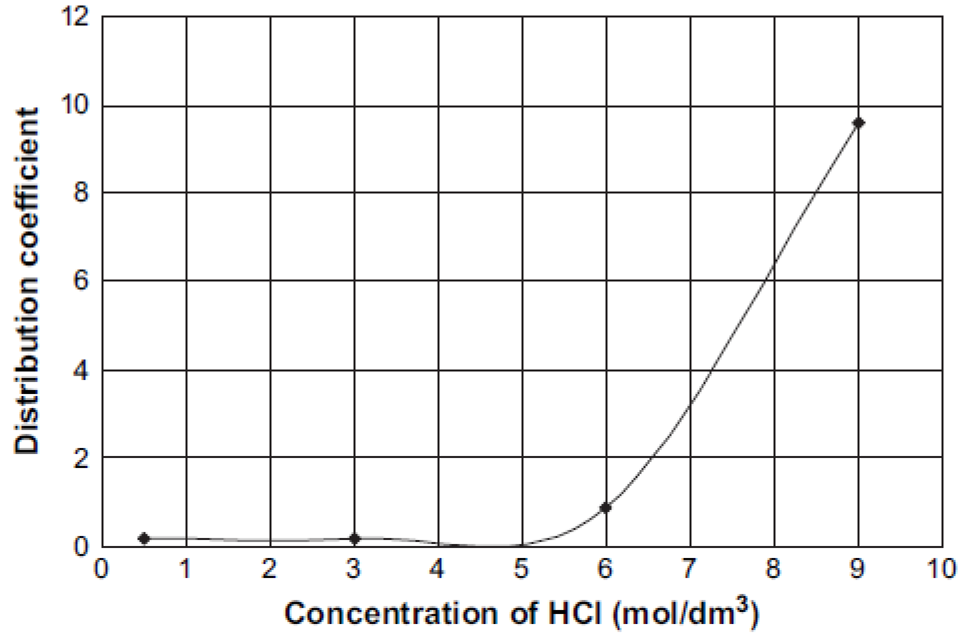


Fig. 4. Distribution coefficient of calcium between benzo-18-crown-6 resin and HCl aqueous solution.

HCl aqueous solution: ε~1.003

× ~3?

Liq./Liq.

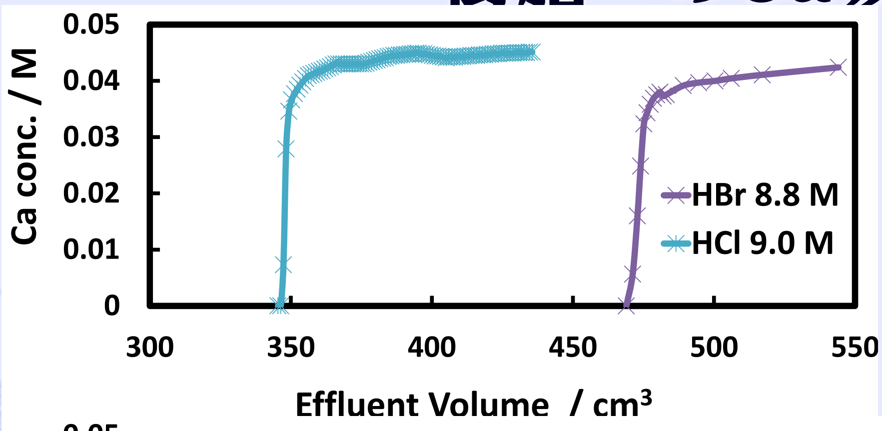
$\varepsilon(\alpha_{40}^{48})$= ？？？ by *pure organic solvent*

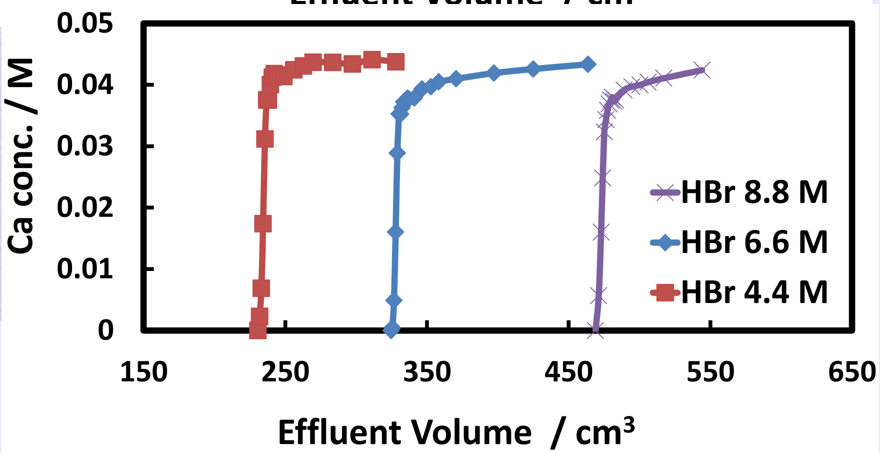
=1.008 by *water-alchohol mixture*

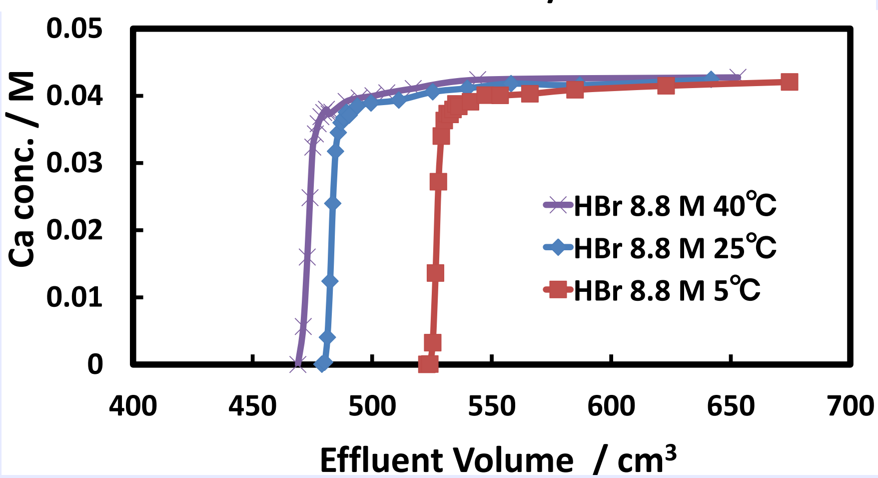
(chloroform-methanol)

Jepson & DeWitt, J. Inorg.nucl.Chem38(1976)1175

樹脂へのCa吸着量の向上







硬い Cl^-＜Br^- 軟らかい

[クラウン − Ca^{2+} − $2X^-$]
アニオン量に依存

温度が低いほうが大きい

佐藤、大井@上智大学＆野村@東工大
原子力学会2013年3月

Prospect for Mass production

Liq./Liq. Extraction

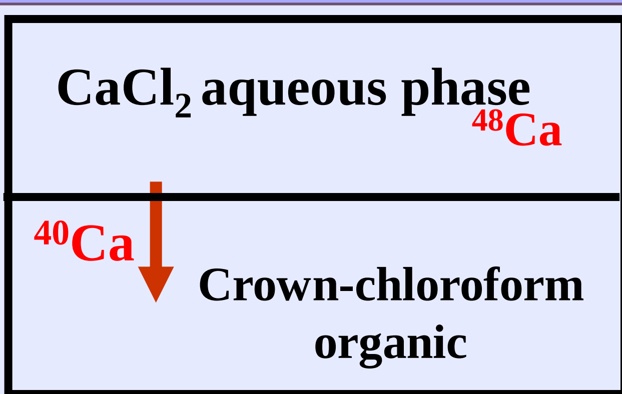
LLE by Microchannel/reactor

- Fast & Highest conversion synthesis
- Aqueous-organic multi-phase flow & process

$$^{40}Ca^{2+}(aq) + {}^{48}CaL^{2+}(org)$$

$$\uparrow \quad \downarrow$$

$$^{48}Ca^{2+}(aq) + {}^{40}CaL^{2+}(org)$$



Solid/Liq.

Column chromatography using crown ether resins

- Multi-stage process
- Slow & low conversion

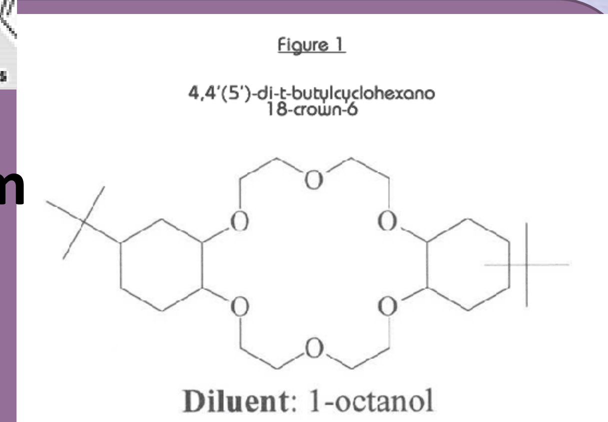
Ca solution: Analyte(mobile phase)

Packed column (stationary phase)

SuperLig樹脂

II

Eichrom or IBC Resin



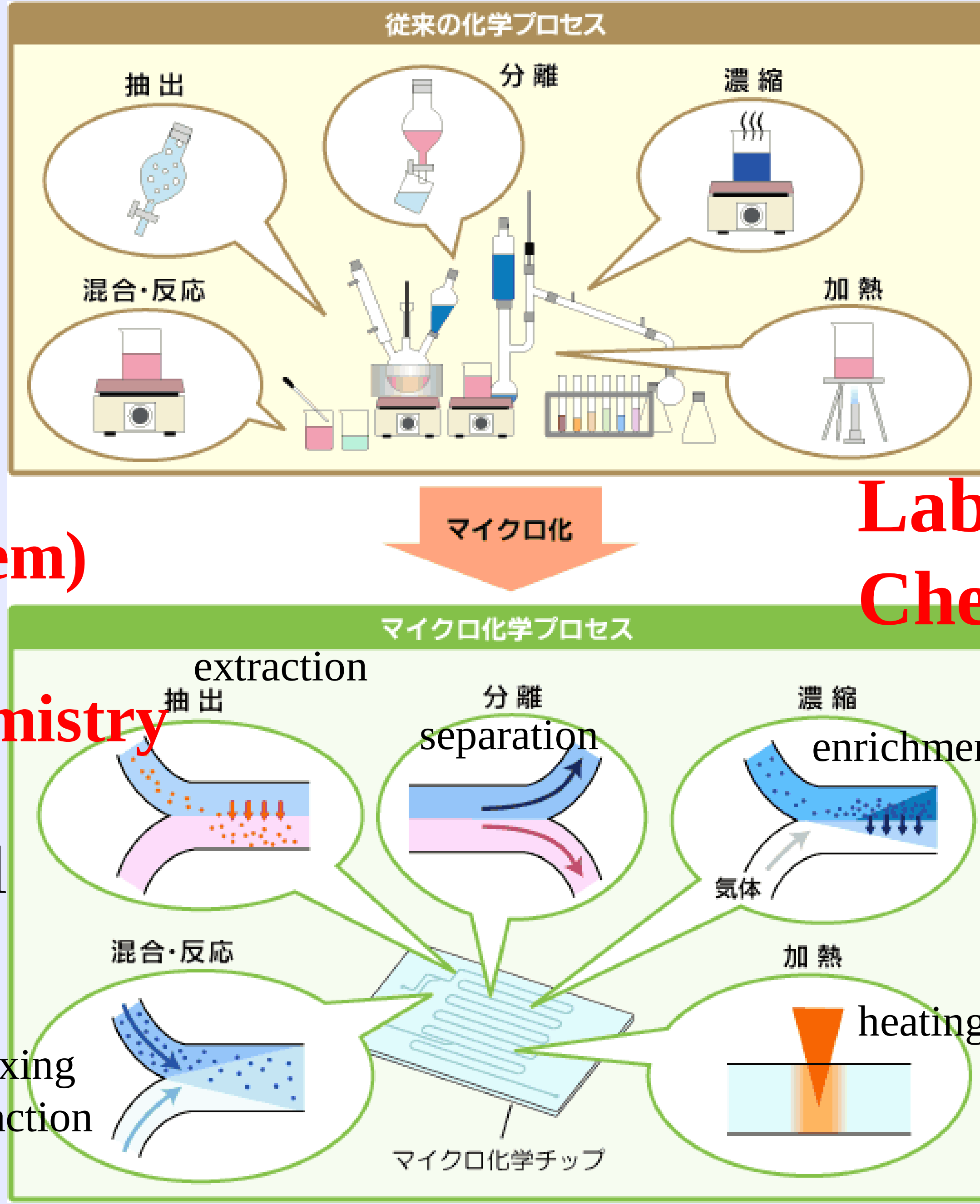
Conventional Chemical Process

μ TAS (Micro Total Analysis System)

&

Synthetic chemistry

Lab-on-a-chip Chemistry



Microchemical Process

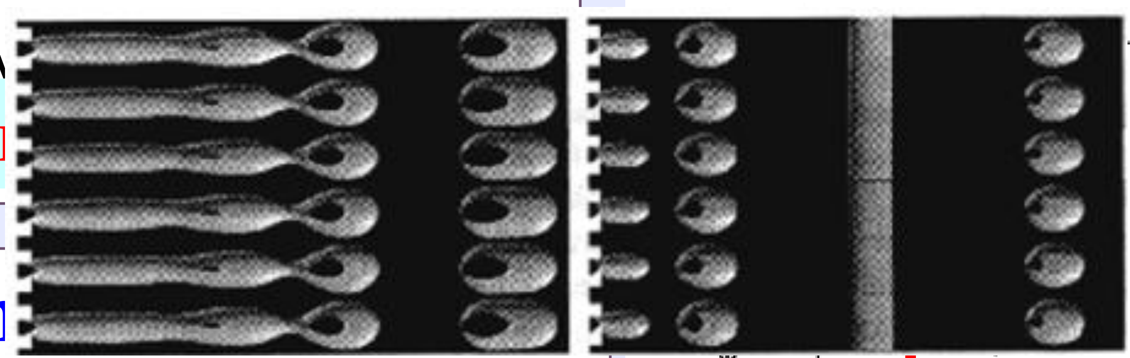
Liq./Liq. extracion → two keywords

Specific interfacial area

(surface to volume ratio:S/V)

S/V = dL/(w

W=100µ



Diffusion

T= **W^2** /D

(D=10^{-9} m^2/s

typical # for molecular in water)

W=100µm → T=**10s** !

(10µm → T=0.1s!)

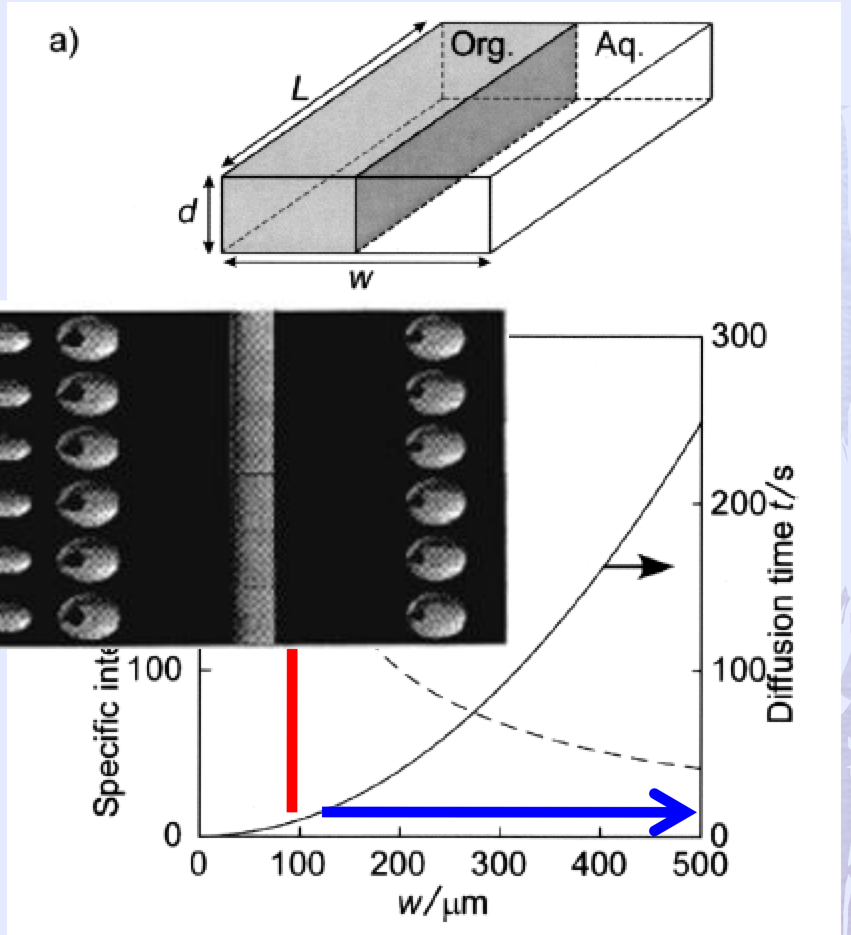
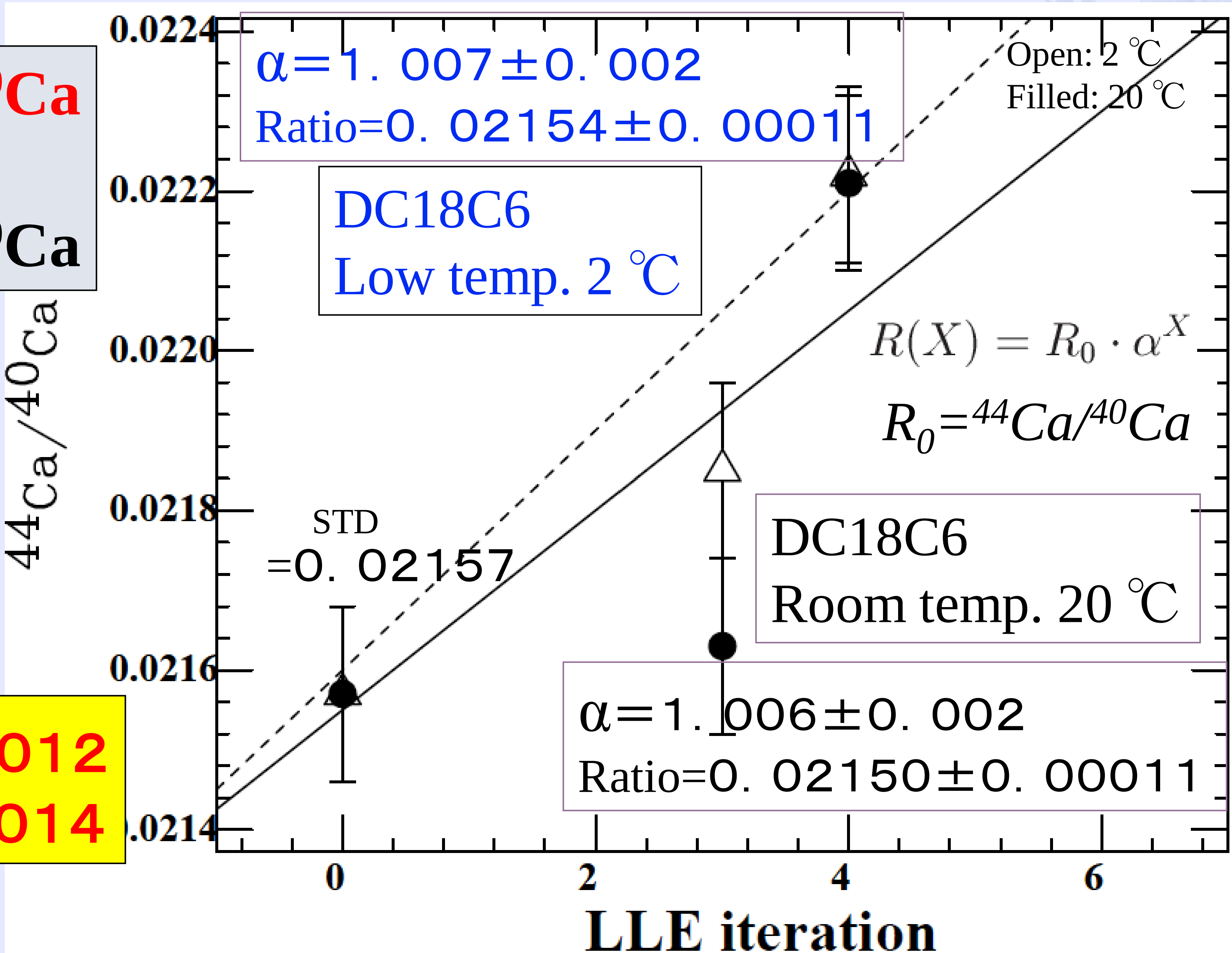


Fig. 2 (a) Illustration of a model of a liquid-liquid interface between the aqueous and organic phases, (b) Specific interface area and diffusion time dependence on the microchannel width

Isotopic Analysis by Reaction-cell ICP@JAMSTEC

$^{48}Ca/^{40}Ca = 2 \times {}^{44}Ca/^{40}Ca$

α～1. 012 ～1. 014



Comparison

Solid./Liq.

- $\varepsilon(\alpha_{40}{}^{48})$=**1.003**
- 200m & 10 days
  → ^{48}Ca×**1.34**
- Absorption of Ca depends on concentration of hydrochloric acid(9M HCl)

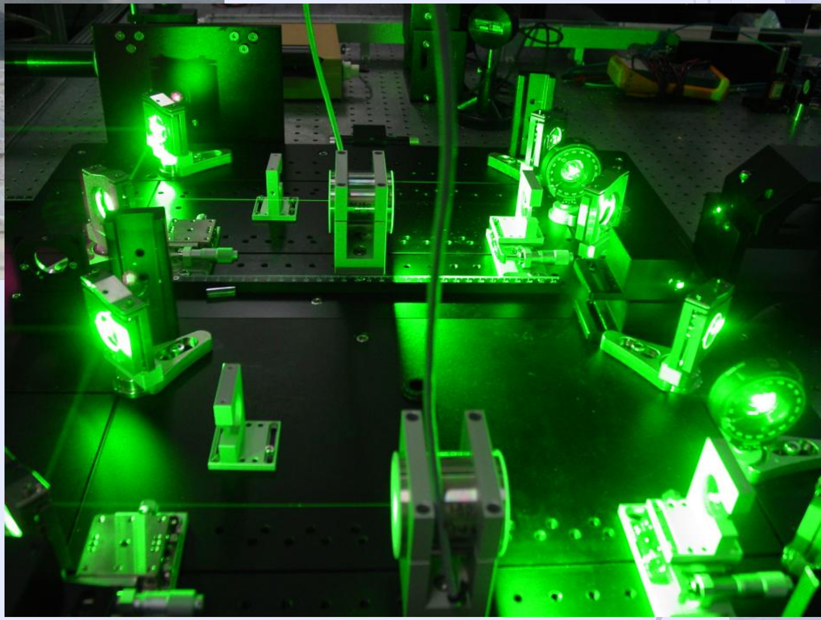
Liq./Liq.

- $\varepsilon(\alpha_{40}{}^{48})$=**1.014:batch**
  ×2 (1.008 by Jepson)
  J. Inorg.nucl.Chem38(1976)1175
- →×1.34 requires ~**20 LLE (~2days)**
- **wide variety/option**
- **Ca concentration**

one order improve! with ~1/6 time:13min by microreactor

KAERI 한국원자력연구원 Korea Atomic Energy Research Institute

KAERI,
Daejeong, Korea
@May, 2008



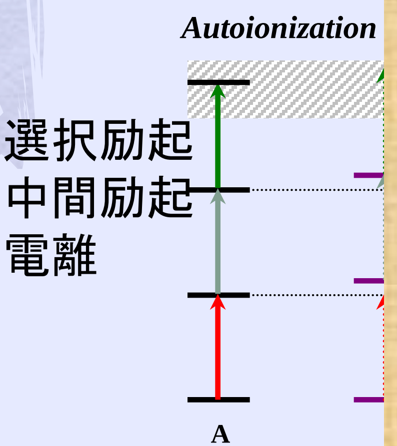
< 70W optical power of 528nm laser >

Int. Conf. on
Laser Application in Nucl. Eng.
April 23 – 25, 2013@ Yokohama

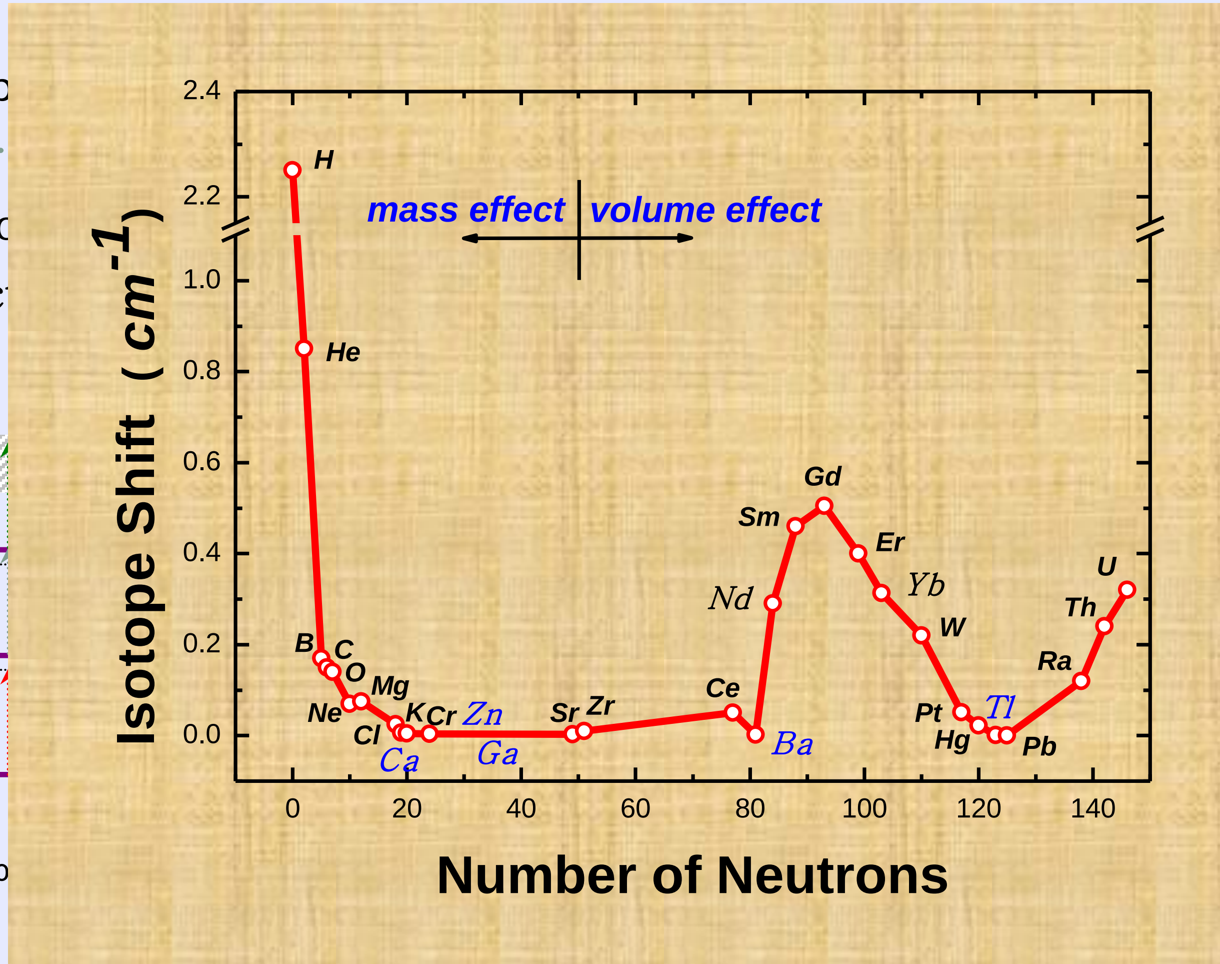
LISOP of Ca-48

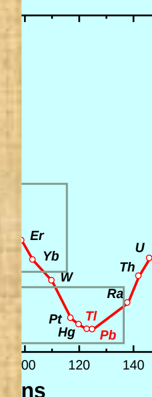
Hard to separate by AVLIS
due to small IS

- AVLIS
  - Multistep p … ade-off)
- LISOP (laser …
  - ISOP (isoto … ectivity.
  - Non-selec…



<3-step photoio…





…ionization states

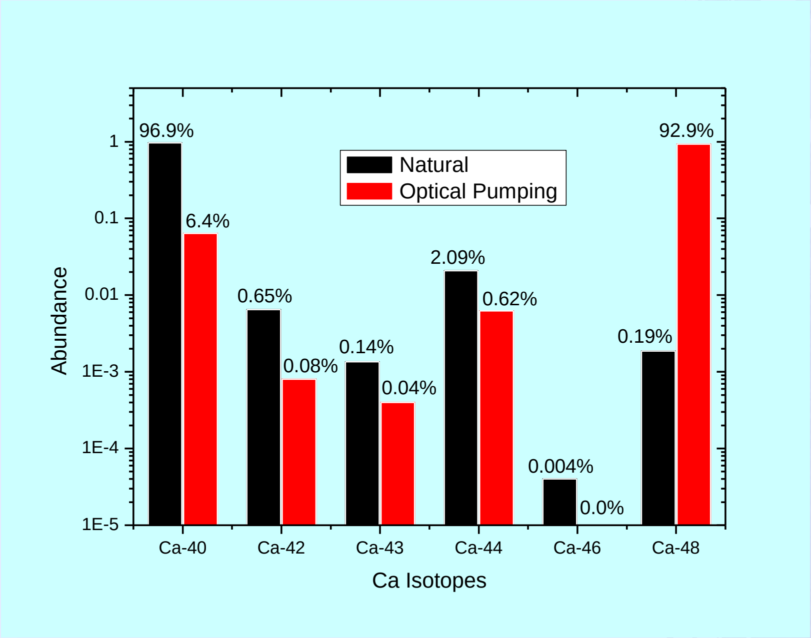
…ck of AIs.

→ ISOP

Do-Young Jeong, KAERI

Application to Ca-48

- Optical Pumping is effective for Ca.
  - Optical Pumping of Ca-48 to >90%
  - Final enrichment higher than 10 % feasible

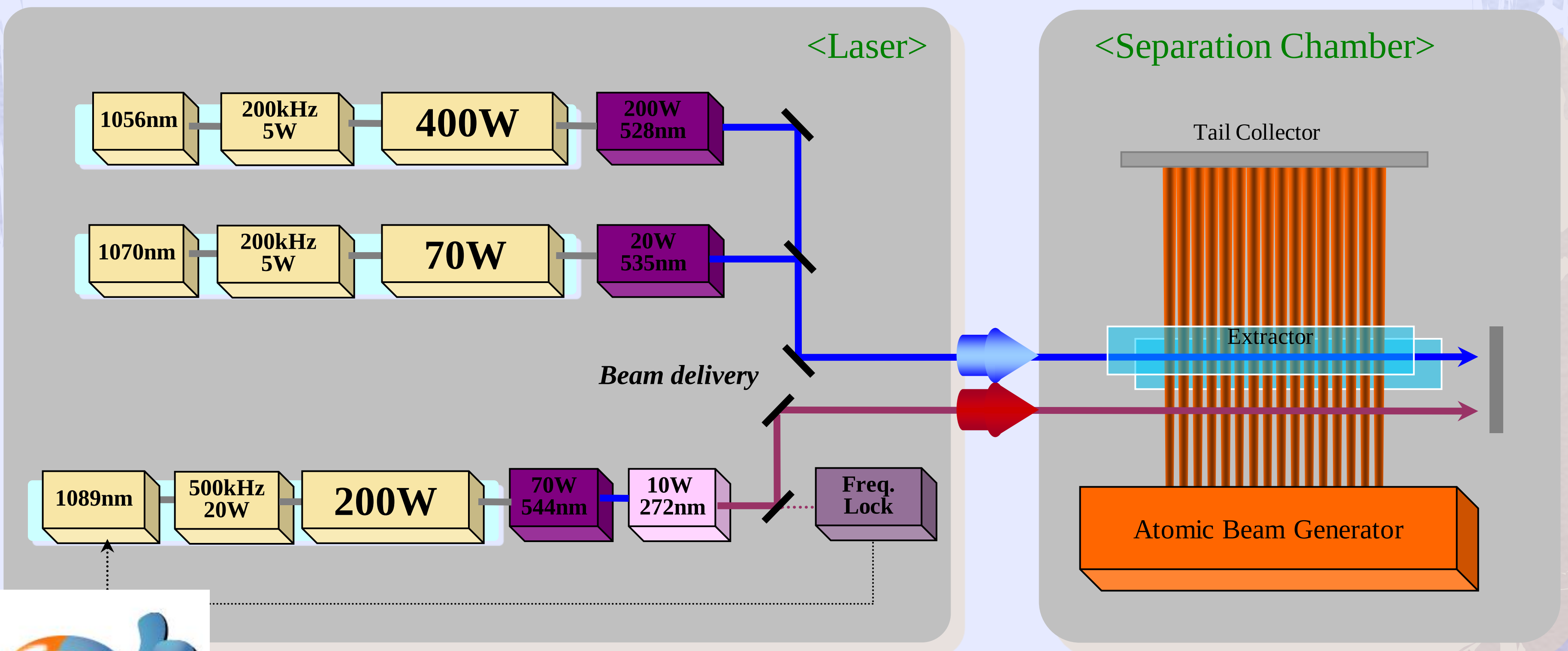


Simulation of Optical Pumping Process

Do-Young Jeong, KAERI

Pilot System

- **Fiber-based laser**
- **Productivity : 1 kg/yr for 20%-enriched ^{48}Ca**



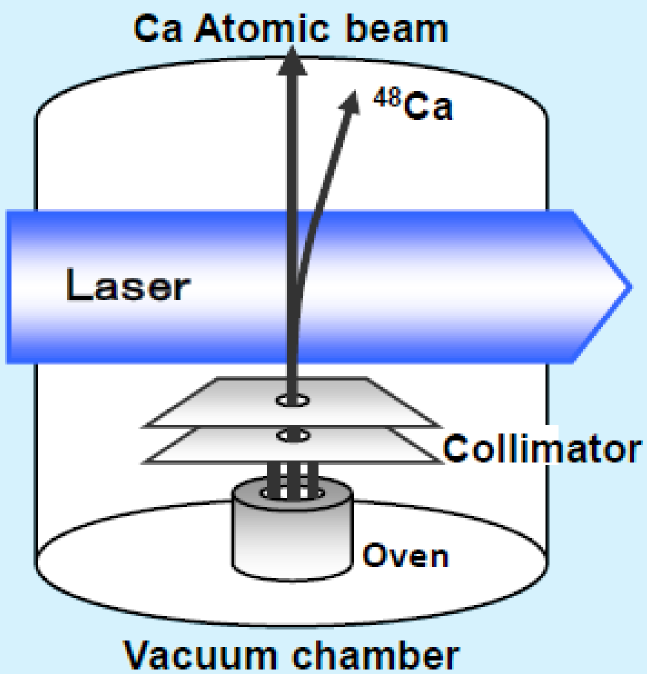
Do-Young Jeong, KAERI

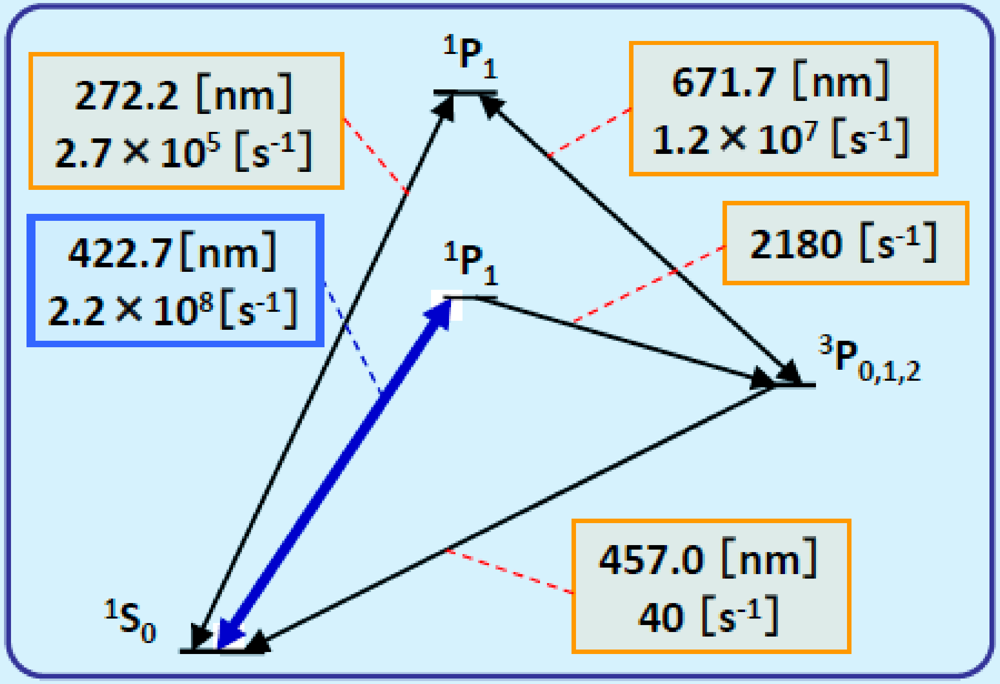
Fiber-based Laser Development

	Requirements	Current status	Remarks
Optical pumping (272 nm)	• 15 ns, 500 kHz • 10 W • $\Delta\nu$ < 250 MHz	• 15 ns, 500 kHz • 5 W • $\Delta\nu$ < 250 MHz	Being improved
Excitation (535 nm)	• 15ns, 150kHz • 20W • $\Delta\nu$ < 5 GHz	• 15ns, 150kHz • 20 W • $\Delta\nu$ < 5 GHz	Completed
Ionization (528 nm)	• 15ns, 150kHz • 200 W • $\Delta\nu$ < 5 GHz	• 15ns, 150kHz • 120 W • $\Delta\nu$ < 5 GHz	Being improved

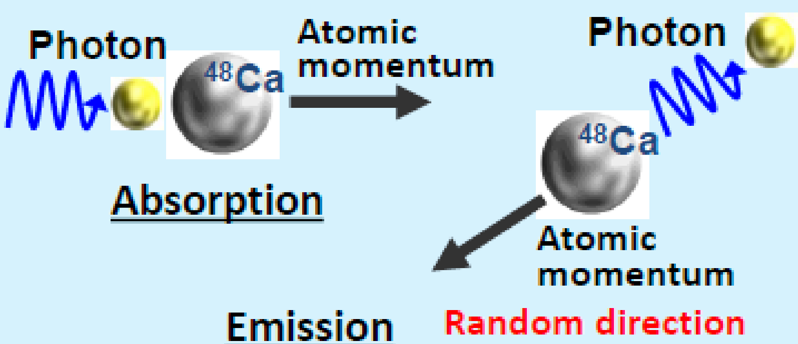
Do-Young Jeong, KAERI

Deflection of ^{48}Ca by Radiation Pressure





Energy levels and Transition Probabilities

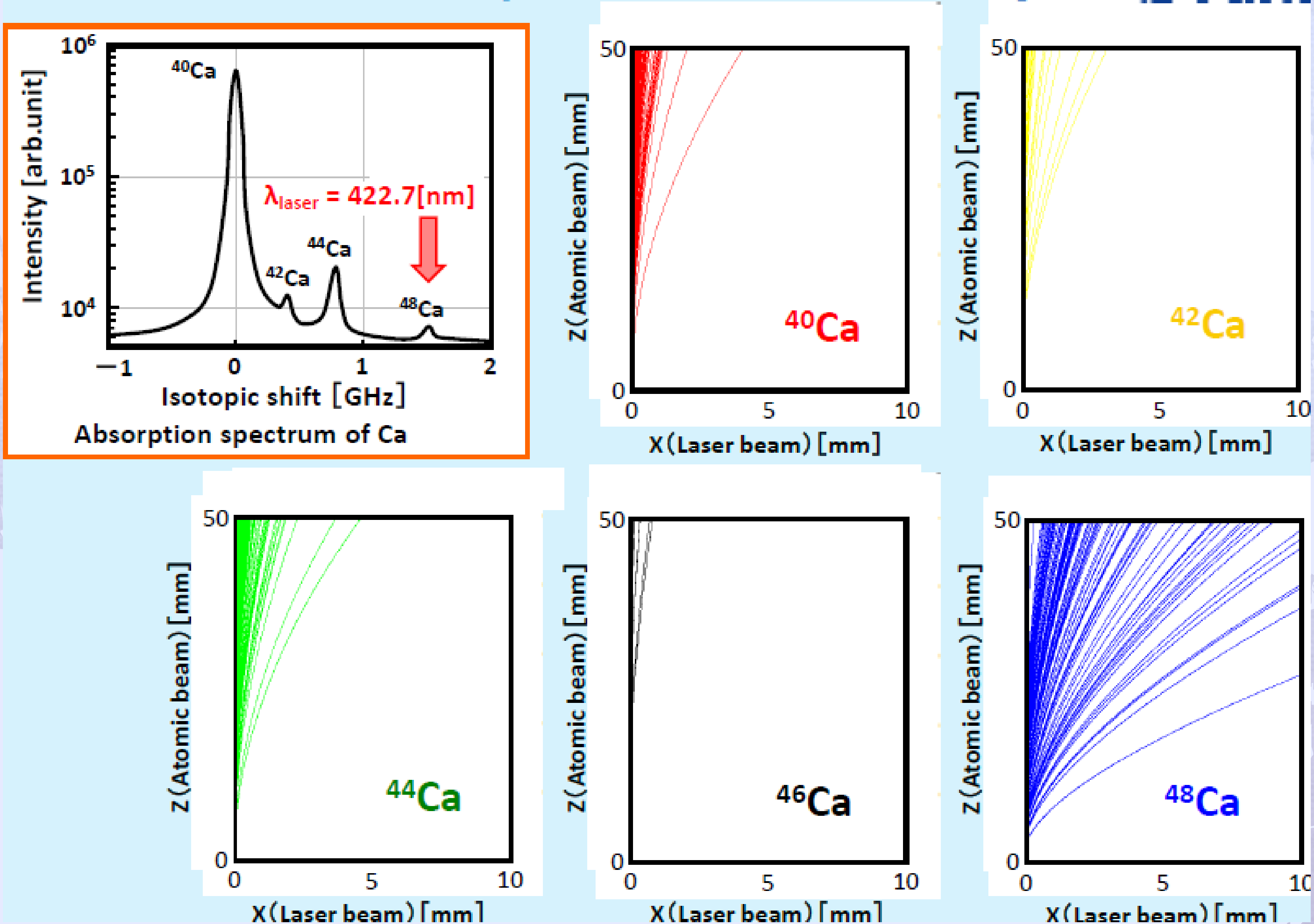


$^{1}P_{1}$ – $^{1}S_{0}$ transition

- Large transition probability
- Quasi Two-Level System

Prof. Niki @Univ. of Fukui

Calculated Trajectories of Ca Isotope



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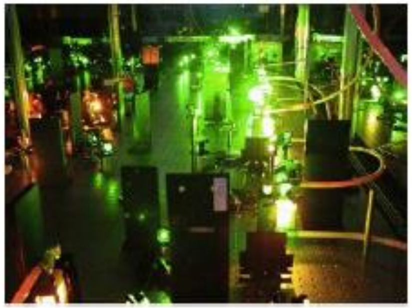
Isotope enrichment

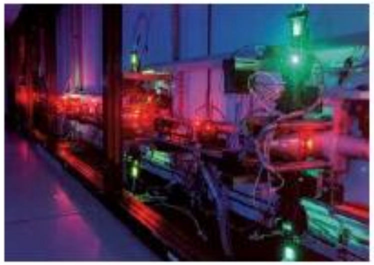
Nucleus	Existing method	R&D
^{48}Ca		Laser separation, gazeous diffusion
^{76}Ge	Centrifugation	
^{82}Se	Centrifugation	
^{96}Zr		Laser separation
^{100}Mo	Centrifugation	
^{116}Cd	Centrifugation	
^{130}Te	Centrifugation	
^{136}Xe	Centrifugation	
^{150}Nd		Centrifugation, Laser

R&D in KAERI (Korea) for ^{48}Ca enrichment by laser



R&D in Russia for ^{150}Nd enrichment by centrifugation

R&D in France for ^{150}Nd enrichment by laser



F. Piquemal, Neutrino 2012 @Kyoto

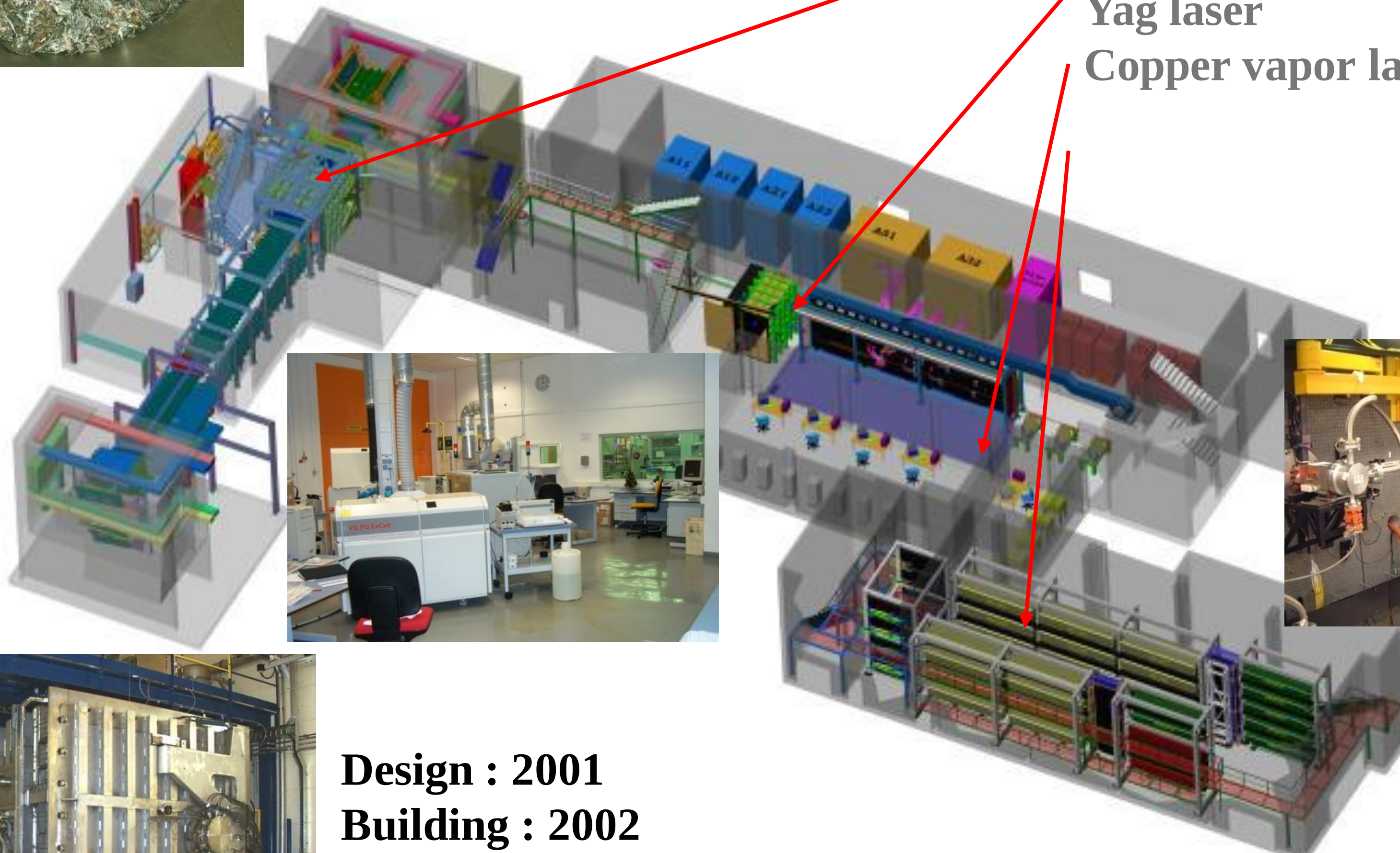
All DBD isotopes produced in Russia up to now

Menphis (Menelas separator) facility

- **Production of 200 kg of enriched U at 2.5 %**
- **About 2000 kg natural U evaporated**

Pierrelatte, France

Evaporator
Dye laser chain
Yag laser
Copper vapor laser



Design : 2001
Building : 2002
1st test : early 2003
1st full scale exp. : june 2003
International collab. '^{150}Nd' created : 2007 SNO+, SuperNEMO

France has abandoned development of AVLIS

- It has begun looking into the implementation of the technology on a commercial scale in 20 yrs

  To get a producation level of kg/hour:

  high power electron gun & high laser power

- CEA wasted money by keeping research on AVLIS going after the technology's lack of feasibility had become apparent

- Nd has been enriched in ^{150}Nd at 60% with a production yield of 40mg/h → ~100 yr for 40kg!

(Kurchatov Institute QE 35(10), 879 (2005)

H. Park et al.(KAERI) : enriched ^{176}Yb at > 97% with about 6mg/h

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Productivity will be an issue!?

Some plants are not flexible(MENPHIS can produce only ^{150}Nd)