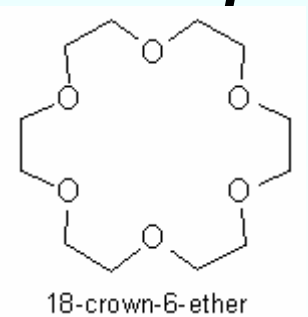
Candles

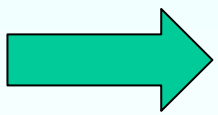
August 9, 2006@6th Recontres du Vietnam, Hanoi

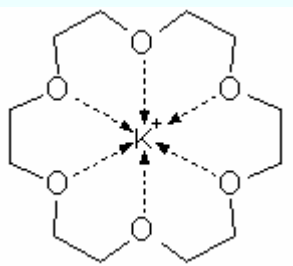
CAlcium fluoride for studies of Neutrino and Dark matters by Low Energy Spectrometer

Challenge on ^{48}Ca enrichment for CANDLES double beta decay experiment

~Separation with a crown ether~







Pedersen@1962
Cram&Lehn@1987
Nobel Prize

Ryuta Hazama

Hiroshima University

広島大学

大阪大學

Double beta decay of ^{48}Ca

- Largest Q value (4.27 MeV)
  - next largest ^{150}Nd (3.3 MeV)
  - Large phase space factor
  - Least background (γ: 2.6 MeV, β: 3.3 MeV)

Isotope	**^{48}Ca**	**^{76}Ge**	**^{82}Se**	**^{100}Mo**	**^{116}Cd**	**^{136}Xe**	**^{150}Nd**
Q-value (MeV)	**4.27**	**2.04**	**3.00**	**3.03**	**2.80**	**2.48**	**3.37**
$G_{0\nu} \times 10^{-25}$ (year^{-1}eV^{-2})	**2.44**	**0.244**	**1.08**	**1.75**	**1.89**	**1.81**	**8.00**

- Next generation
  - $M_\nu \propto 1/T^2 \propto M^2$ if background free
  - $M_\nu \propto 1/T^2 \propto M^4$ if background limited
- ^{76}Ge experiment (already seen backgrounds)
- ^{48}Ca (no backgrounds seen) large Q value

CANDLES III

CaF_2(pure) : $10 \times 10 \times 10 cm^3$

60 Crystals (191kg)

Liquid Scintillator

Wave Length Shifter

4 π Active Shield

UV ↓ visible

Outside View

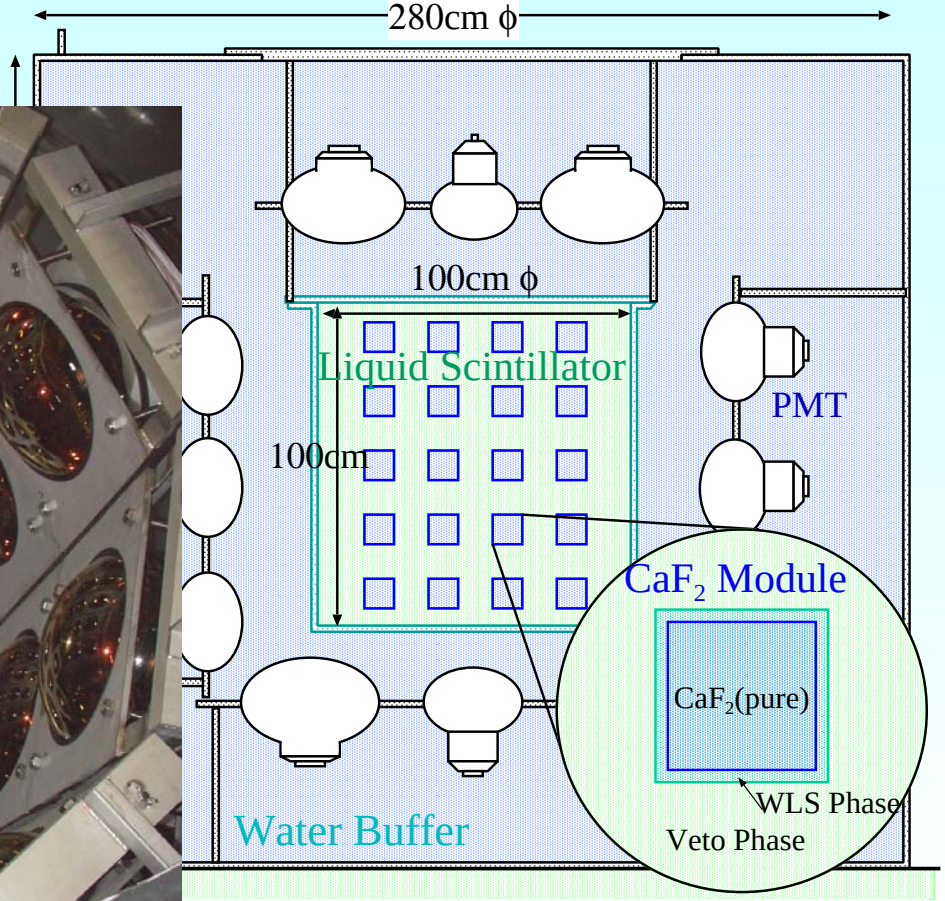
Inside View

40 PMTs

15” PMT (×8) : R2018

13” PMT (×32) : R8055

Water Tank for CANDLES III



Different time constant

Undoped CaF_2 ~1 μ s

Liquid Scinti.~a few 10ns

Mile stone

NPA 730 '04, 215

$T_{1/2}^{0\nu\beta\beta} > 4.5\times10^{22}$ year (68 % C.L.)

$> 1.4\times10^{22}$ year (90 % C.L.)

$\langle m_\nu \rangle < 7.2 \sim 44.7$ eV (90 % C.L.)

- ELEGANTS VI
  - World Best Value(^{48}Ca:6.4g)
- CANDLES I, II (proof of principle;4 π active&FWHM~4%@Q)
- CANDLES III (construction completed@ Osaka lab.)
  - CaF_2(10cm^3) 200kg:sea level, 300 kg; Kamioka
  - ~30 μBq/kg for ~0.5 eV

achieved

- CANDLES IV
  - 15cm^3 cube (600 crystals) 6.4t(^{48}Ca:6kg); Kamioka
  - ~3 μBq/kg for ~0.1 eV in 6 years
- CANDLES V
  - 100t; SNO or Kamland or … for ~30meV in 7years

^{48}Ca enrichment

- Natural abundance

  → 0.187%

  – Enriched isotope

  → expensive (~~elemag. separator~~;calutrons)

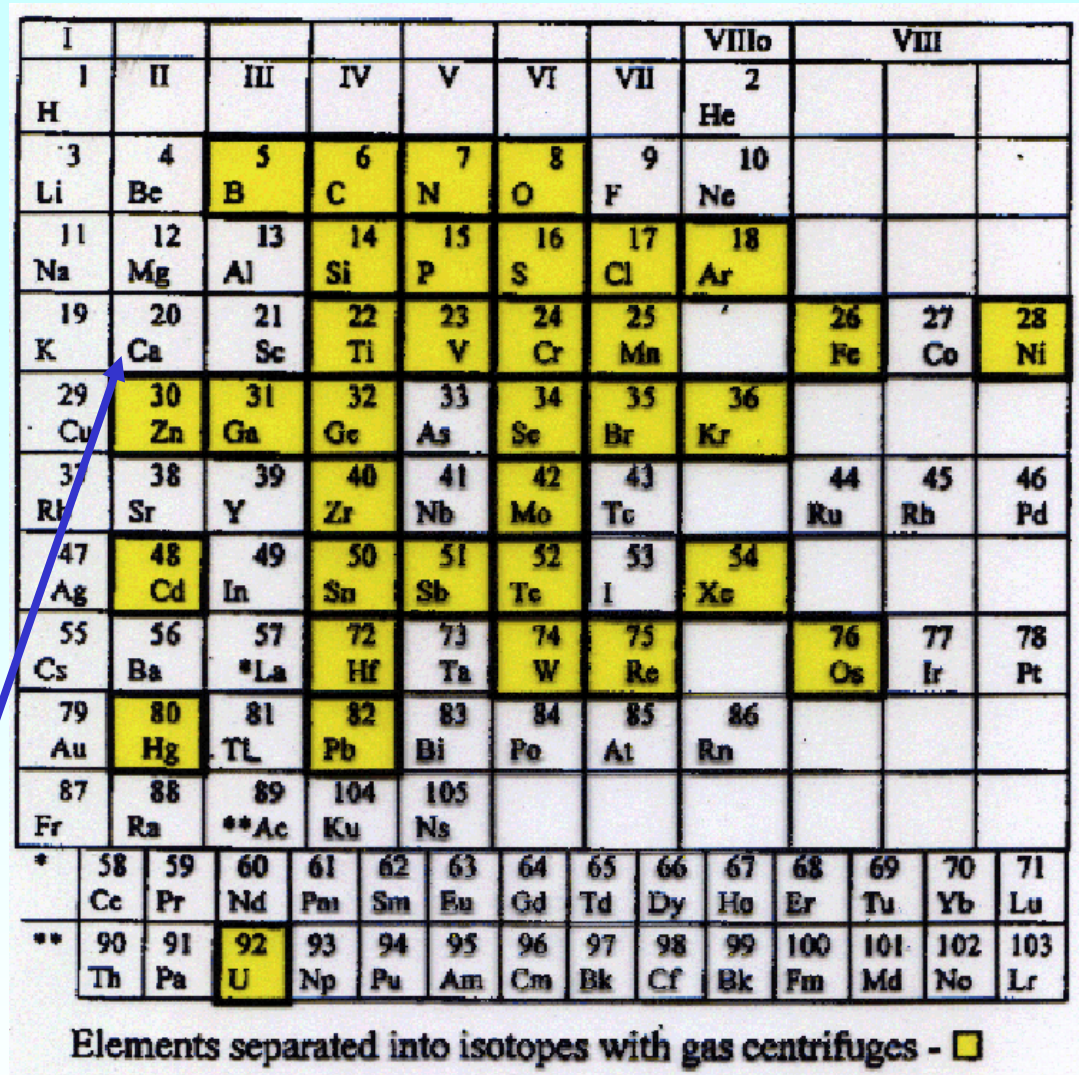
  ~200K$/g

  ~10g × 2 (in the world)

  →no gaseous compounds

  at room temp.

  ~~Gas centrifuge~~

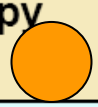


A.I.Karchevski

ββ isotopes; ^{48}Ca, ^{96}Zr, ^{150}Nd etc.

Technologies for isotope production

for Ca

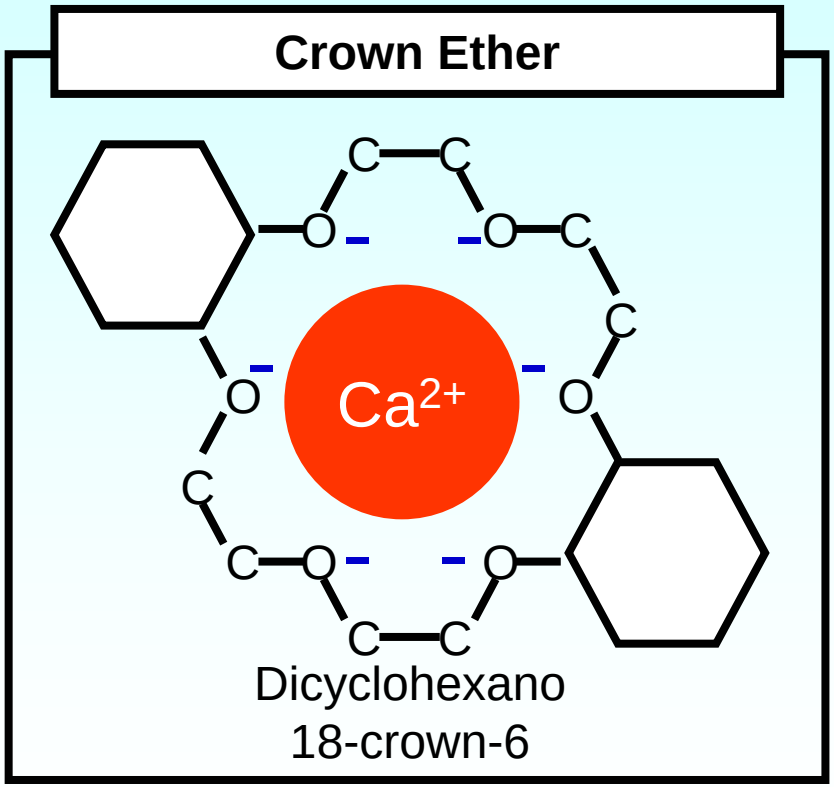
Separation technology	*Field of use*	*Production per year*	*Cost*
Electromagnetic (mass-spectroscopy effect) 	universal	tens of grams	high
Chemical & phys. processes (rectification, chem. exchange etc) 	light elements	tons	low
Gas diffusion 	elements forming gas compounds	thousands of tons	middle
Gas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

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

Unique Property of Crown Ether

Complexing of cations(anions) by neutral molecules is an uncommon phenomenon.

Stability is ~10^4 × no-ring(crown)

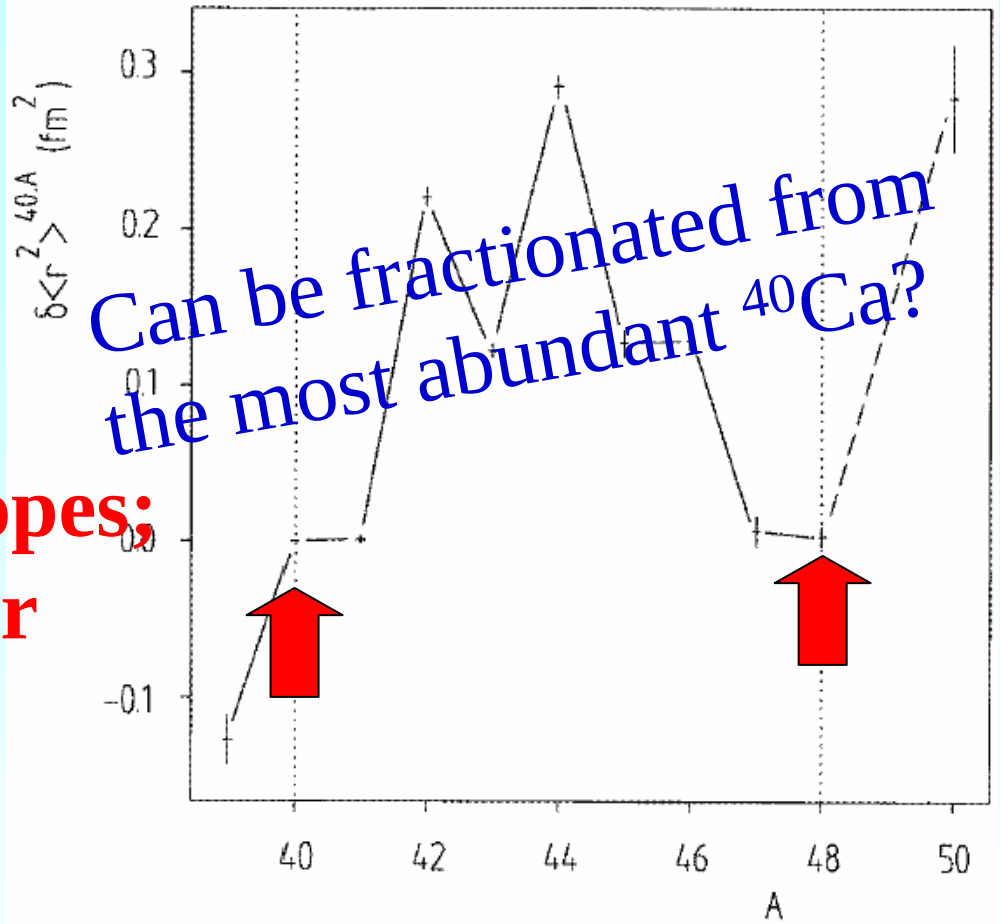


- **Held by electrostatic attraction between negatively charged O^- of the C-O dipoles & cation (Ca^{2+})**
- **How well the cation fits into the crown ring**
- **Liquid(aq-salt)-liquid(org-crown) extraction in isotopic equilibrium**

DC18C6

- Total # of atoms in the ring
- # of oxygen atoms in the ring

The mean square Nuclear charge radius of Ca



Two doubly magic isotopes; A parabolic behavior

L.Vermeeren et al., J.Phys.G,22(1996)1517

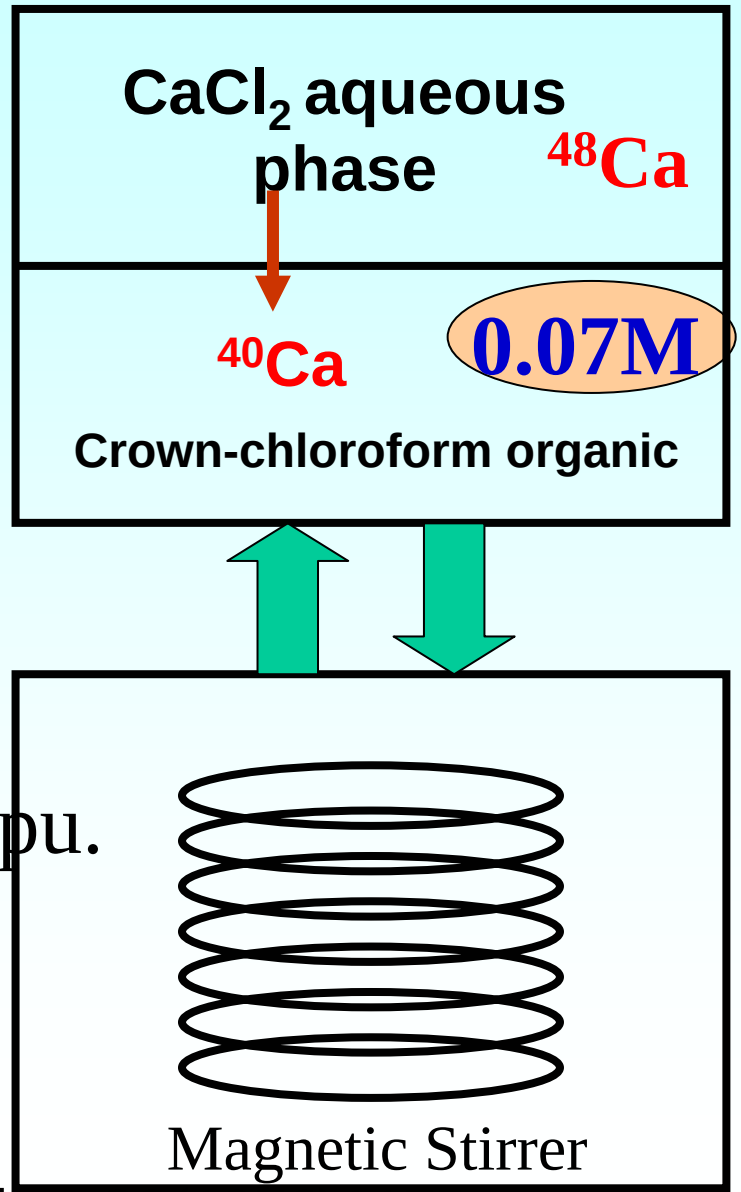
Ca isotope	^{40}Ca	^{42}Ca	^{43}Ca	^{44}Ca	^{46}Ca	**^{48}Ca**
abundance (%)	96.9	0.65	0.135	2.09	0.004	**0.187**

Ca Isotope effects～Separation Principle

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

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

DC18C6: Aldrich Chemical, 98.0%
$CHCl_3$:Nakarai Tesque, 99.0%
$CaCl_2$:Nakarai Tesque, 95.0%



Solvent Extraction process

1.vacant extraction to reduce impu.
2.mixed & stirred for 1 hour
3. standing for 1 hour @7°C
4. LLE iterated 6 times

B.E.Jepson&R.Dewitt, J. Inorg.nucl.Chem38(1976)1175

Major background molecular ions formed from the Ar Plasma, nebulized water and dissolved/contained air.

m/z=

Mass	Molecular ion	isotopic ratio(%)	required resolution
40	$^{40}Ca^+$	99.941	-
40	$^{40}Ar^+$	99.6	192498 ✗
42	^{42}Ca	0.647	
42	$H_2^{40}Ar$	99.57	2162
43	^{43}Ca	0.135	-
43	$^{86}Sr^{2+}$	9.86	10392
43	$^{42}CaH^+$	0.6469	5597
43	$^{40}Ar3H$	0.0298	1683
44	^{44}Ca	2.086	-
44	$^{88}Sr^{2+}$	82.58	16448 ✗
44	CO_2	98.43	1280
44	$^{14}N_2^{16}O$		
48	^{48}Ca	0.187	-
48	^{48}Ti	73.8	10457
48	$^{36}Ar^{12}C$	0.333	2447

Max resolution=12000

Enemy

How to measure ^{40}Ca?

1. TIMS(TRITON Thermo Electron)
   No-Ar

Only four TRITONs in Japan

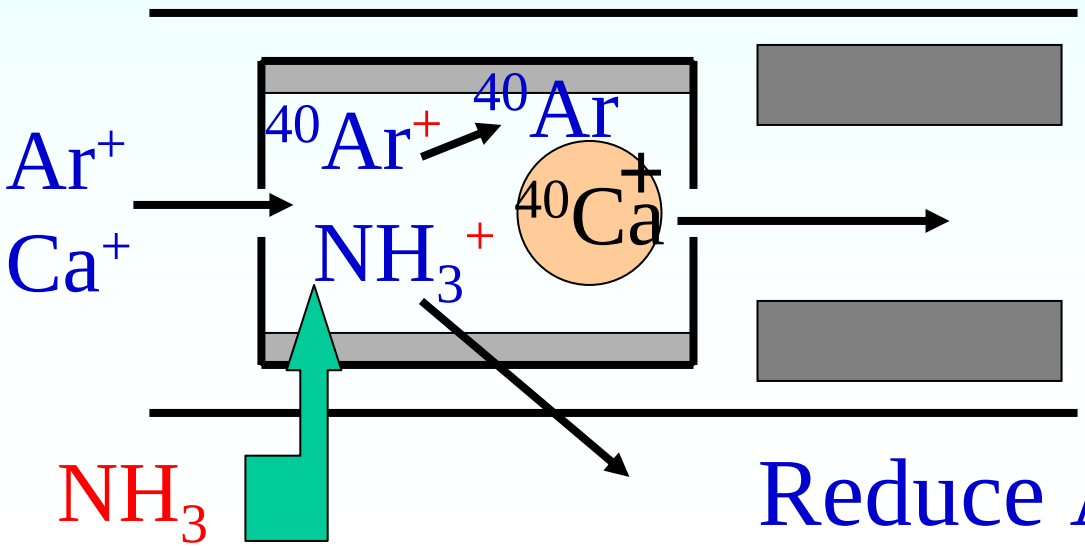
2. Reaction(collision)-cell ICPMS

Perkin Elmer ELAN-DRCII@Kochi Univ.

Q inside reaction-cell allows use of ammonia

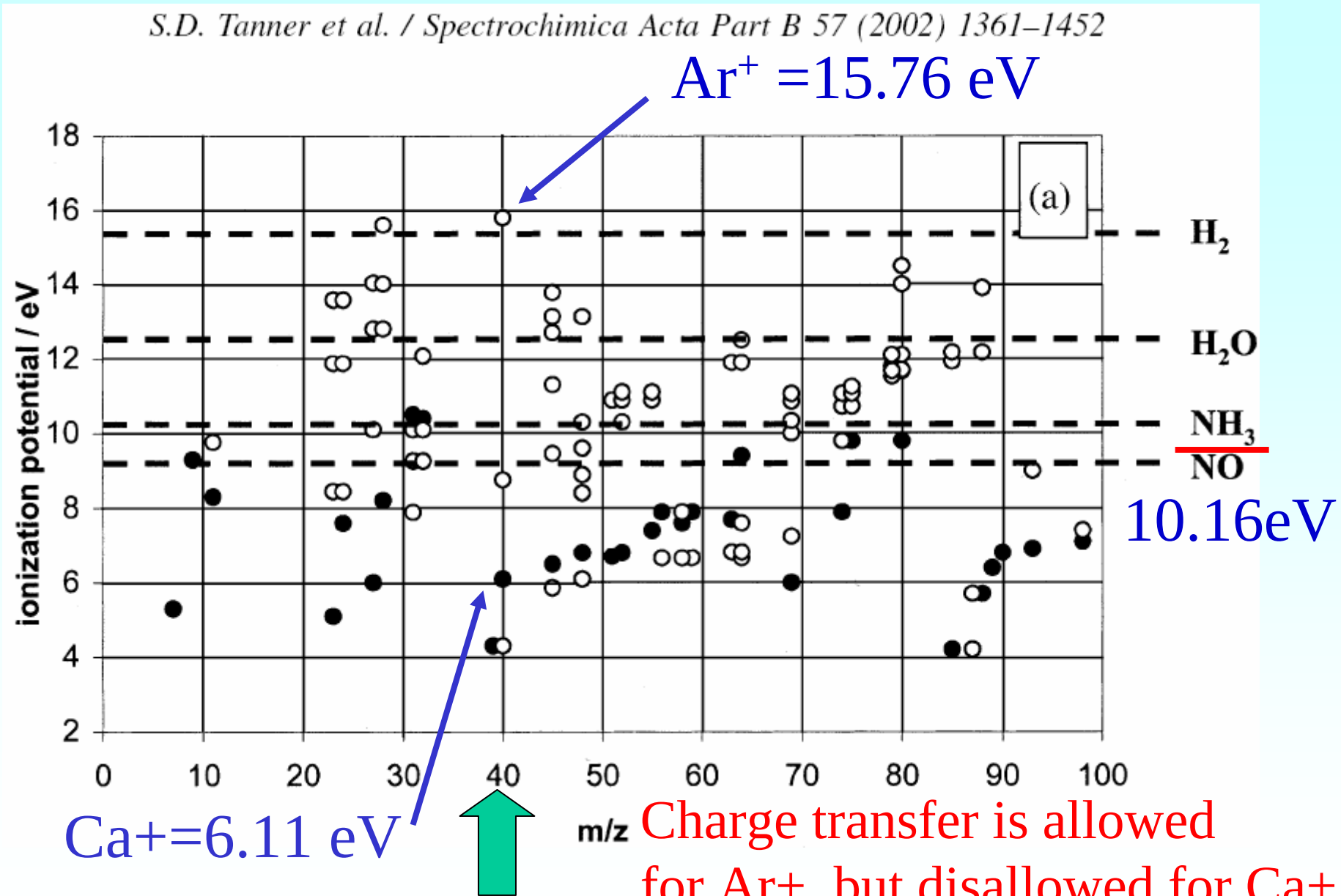
→ can avoid interference of Ar by reaction-gas

Simple collision-cell must use simple gas(H_2, He) to limit adverse side reaction products

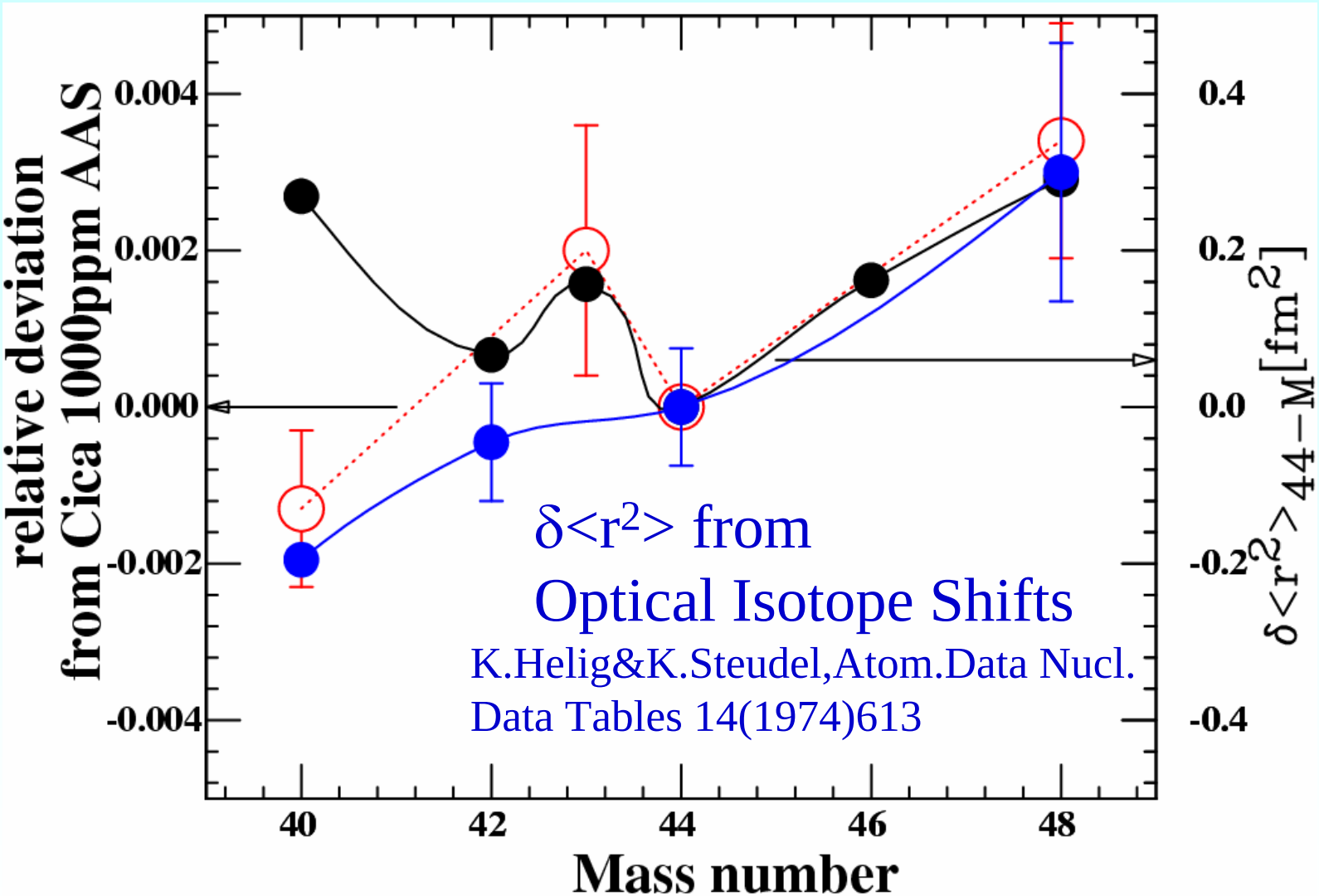


$Ar^+ + NH_3 \rightarrow NH_3^+ + Ar$

Reduce $Ar^+ \rightarrow 10^9 > 10^3$


S.D. Tanner et al. / Spectrochimica Acta Part B 57 (2002) 1361–1452
Ar+ =15.76 eV
(a)
H2
H2O
NH3
NO
10.16eV
ionization potential / eV
m/z
Ca+=6.11 eV
Charge transfer is allowed
for Ar+, but disallowed for Ca+
Ca(6.11eV)<NH3(10.16eV)<Ar(15.76eV)

^{40}Ca,^{48}Ca are doubly magic→ **A parabolic behavior**



Nuclear mass effect > Nuclear size&shape effect!!!

This is crucial asset to realize ^{48}Ca enrichment (from ^{40}Ca)

How small?

Evaluate each isotope effects by 3 measured $\varepsilon(=1-\alpha)$

$\varepsilon_{40\text{-}48}$, $\varepsilon_{43\text{-}48}$, $\varepsilon_{44\text{-}48}$

Bigeleisen theory

$$\varepsilon_{43\text{-}48}=a(\Delta M/MM')_{43\text{-}48}+b\delta\langle r^2\rangle_{43\text{-}48}+(\ln K_{hf})_{43}$$

Nuclear mass effect Nuclear size&shape effect Hyperfine splitting(spin)

	$CaLCl_2$	$SrLCl_2$	$CrLCl_3$
	$b\delta < r^2 > /[a(\triangle M/MM')]$		$lnK_{hf}/[a(\triangle M/MM')]$
$^{40}Ca-^{48}Ca$	0.02 ± 0.48		field shift effect is small!
$^{44}Ca-^{48}Ca$	0.62 ± 1.31		-
$^{43}Ca-^{48}Ca$	0.22 ± 0.88		0.64 ± 1.35
$^{50}Cr-^{52}Cr$	1.12 ± 2.79		almost identical-effect
$^{54}Cr-^{52}Cr$	-2.81 ± 5.97		-
$^{53}Cr-^{52}Cr$	-2.05 ± 8.94		-0.83 ± 6.17

If the field shift effect is dominat, this method is not effective for Ca.

Comparison

Table 1: Summary of previously achieved(measured:known) calcium enrichment. LLC(liquid-liquid chromatography), DC18C6((polyether) dicyclohexyl 18-crown-6), HDEHP(di(2-ethylhexyl) orthophosphoric acid), SLC(solid-liquid-chromatography), LIS(laser isotope separation), MCIRI(Magnetic Cyclotron Ion Resonance of Isotopes)

	separation factor	process	ref.(manufacturer)
1.0020	1.012±0.005 (α_{42}^{48})	LLC(DC18C6)	Osaka RI-center and WERC
1.0028	1.014±0.006 (α_{43}^{48})	LLC(DC18C6)	Osaka RI-center and WERC
1.0010	1.0080±0.0016† (α_{40}^{48})	LLC(DC18C6)	[1]
1.0007	1.0029±0.0006 (α_{44}^{48})	LLC(HDEHP)	[2]
	1.0013±0.0003	LLC(amalgam(Hg))	[3]
	1.000043∼1.000034	SLC(ion-exchange)	[4]resin(Dowex)
	1.00026 (α_{40}^{47})	SLC(ion-exchange)	[5]resin(Dowex)
	1.00021 (α_{40}^{44})	SLC(ion-exchange)	[6]resin(Dowex)
	1.00087±0.00008 (α_{40}^{48})	SLC(ion-exchange)	[7]$NH_4\alpha$-hydroxyisobutyrate&(Dowex)
1.0010	1.0041±0.0004 (α_{40}^{44})	SLC(ion-exchange)	[8] iminodiacetate&resin(ANKB-50)
	1.00013∼1.00087‡	SLC(ion-exchange)	[9](TIT)resin(PK-1),Counter-Current
	1.00016∼1.00037 (α_{40}^{48})	SLC(ion-exchange)	[10](Sophia) resin(Asahi LS-6)
	1.00018 (α_{40}^{48})	SLC(ion-exchange)	[11]resin(AG50WX4)
	1.00049∼1.00013 (α_{40}^{44})	SLC(ion-exchange)18C6	[12]resin(AG50WX4)
1.0010	1.0039±0.0002 (α_{40}^{44})	SLC(cyptand2_B.2.2)	[13]
1.0006	1.0025±0.0003 (α_{40}^{44})	SLC(18C6)	[13]
	1.00011±0.00003 (α_{40}^{44})	SLC(iminodiacetate)	[13]
1.0009	1.0035±0.0003 (α_{40}^{44})	SLC(18C6+dimethylsulfoxide)	[14]
1.0006~1.0013	1.0045∼1.0104(α_{40}^{48})§	SLC(cryptand2_B.2.2)	[15]
	-	LIS(LLNL)	a few $/mg(¥1M/kg) for ^{48}Ca [16]
	20%	MCIRI	5kg/day→10g/day(0.7K$/g)* [17]
	65.3∼95.7%	carbonate or oxide	TRACE Science Int. [18]
	6%% (α_{40}^{44})	chemical diffusion‡	[19]

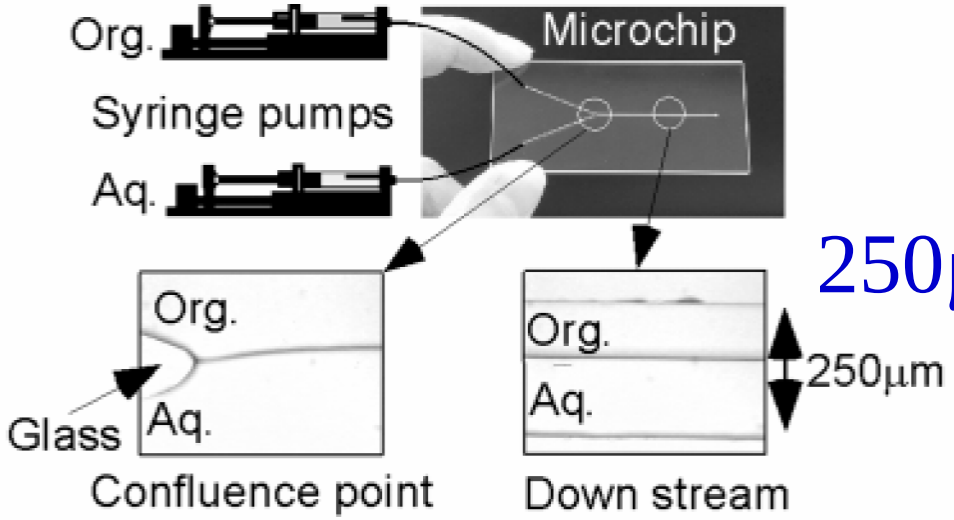
† 0.185% →10% for 1kg/yr by Counter current distribution method.
‡ 0.185% →0.226%‡ after 5 weeks, yielding 144mg of the enriched calcium(1.4g/yr).
§ In a preliminary experiment, they could isolate 30mg of calcium in which ^{48}Ca was enriched by 3.3 % at 0°C from 210mg of natural abundant calcium.
* This corresponds to 3.7kg/yr(¥0.7M/kg). Current cost of product at "electromagnetic"(aka calutrons at ORNL) separation ∼200K$/g(¥200M/kg) .

Preliminary

Need to verify by precise TIMS & More iterate LLE

~800 iteration

0.187→ 2.0%



Macro)10cm cube
S/V~0.6/cm
250μm wide,100μm deep,
and 3cm length
S/V~80/cm

Fig. 1 Photographs showing glass microchip and liquid–liquid interface formed inside the microchannel.

No-stirring,Fast!!

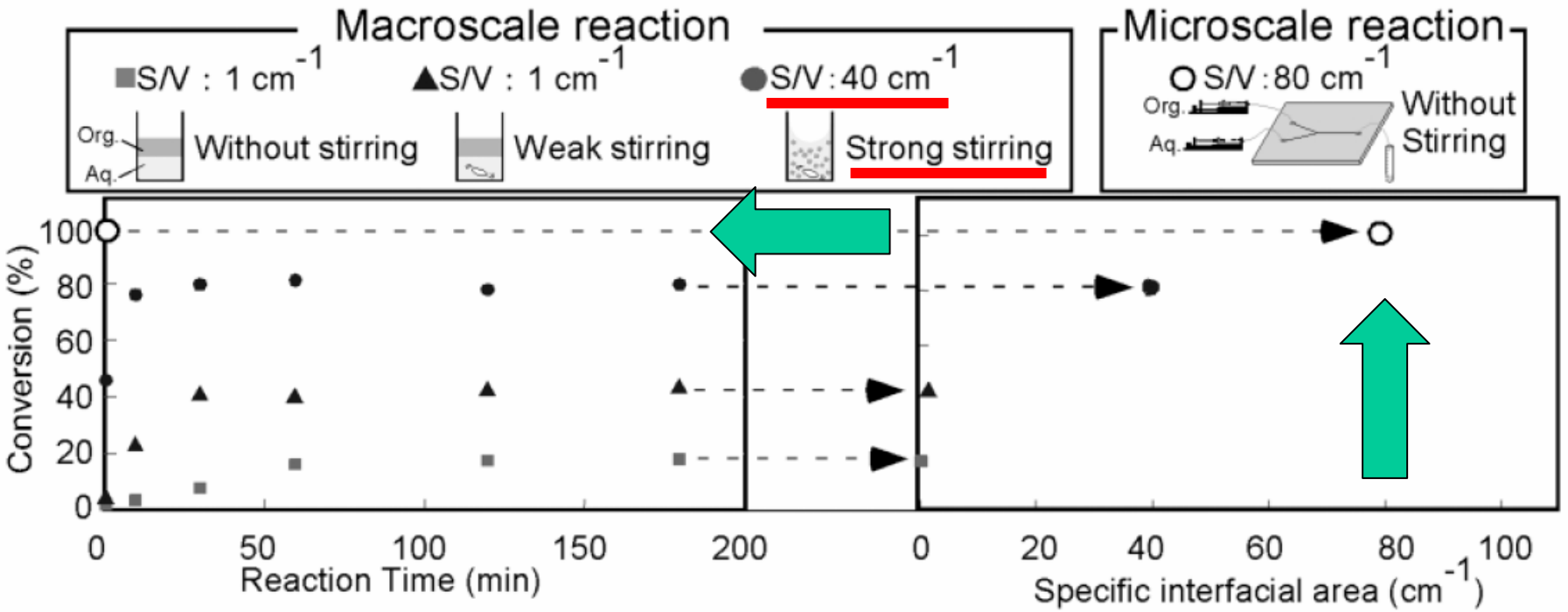
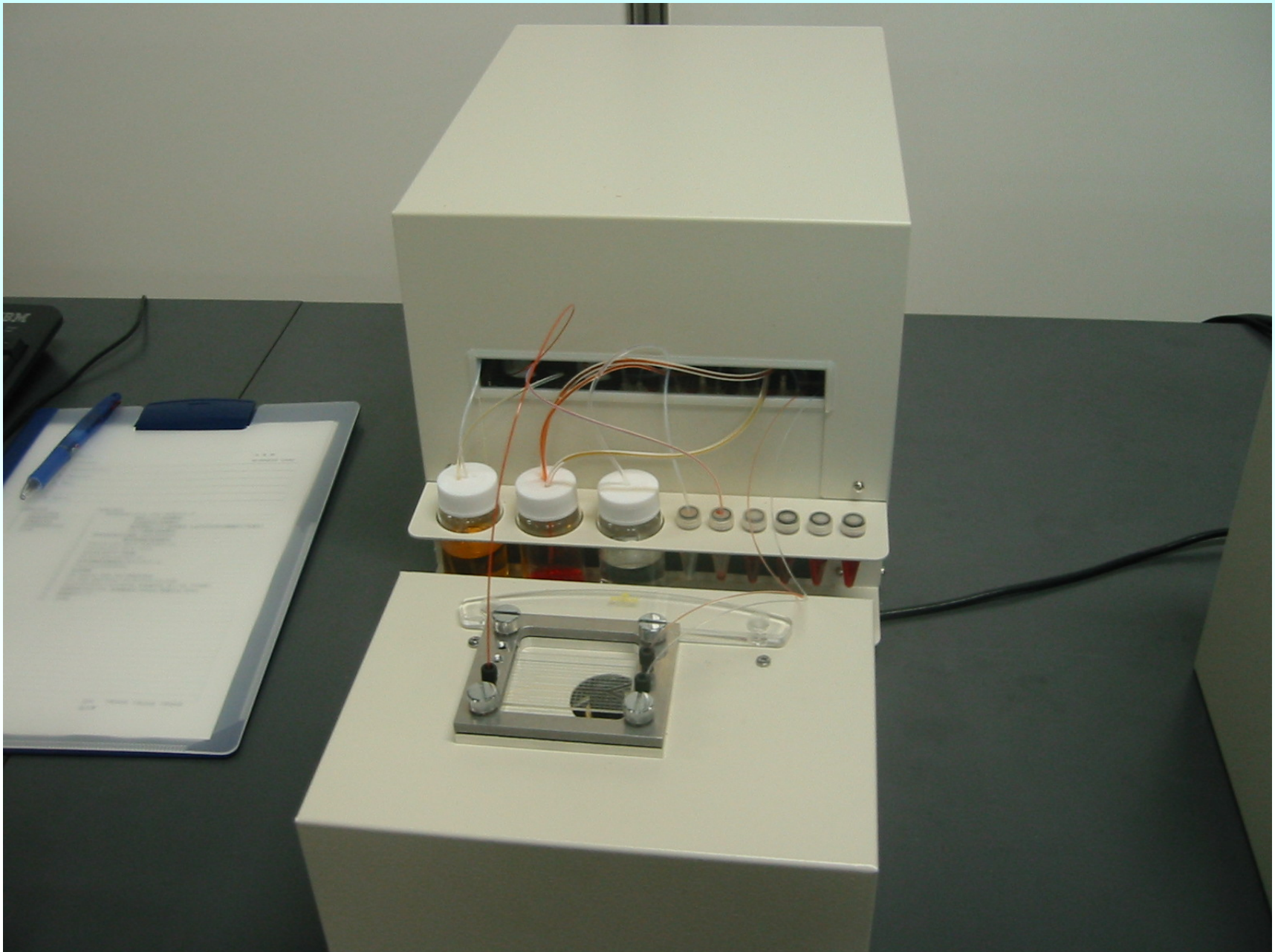
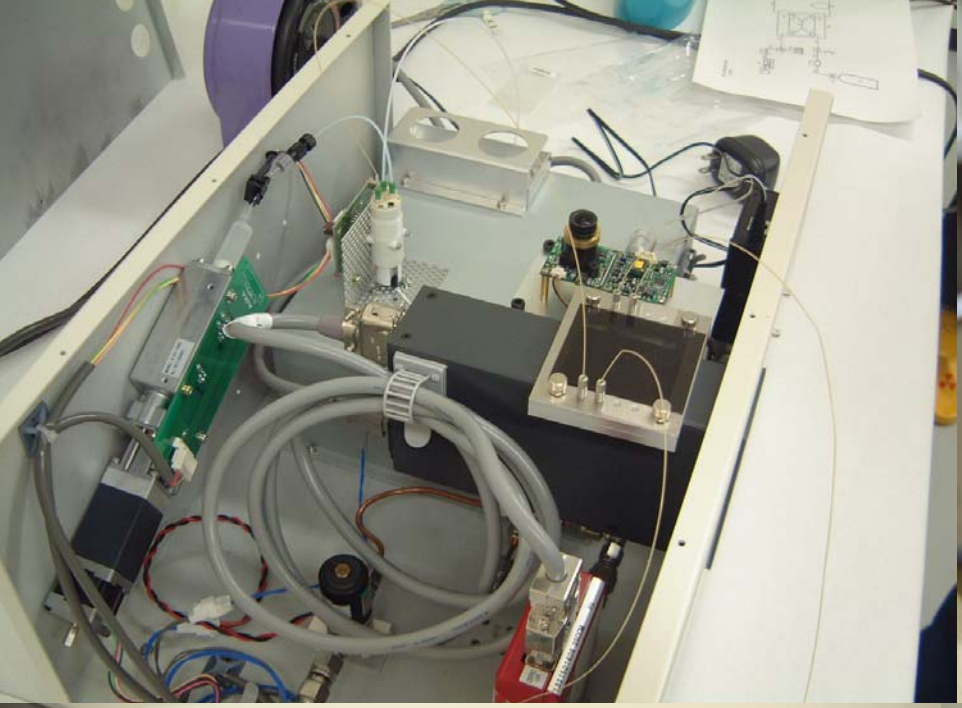
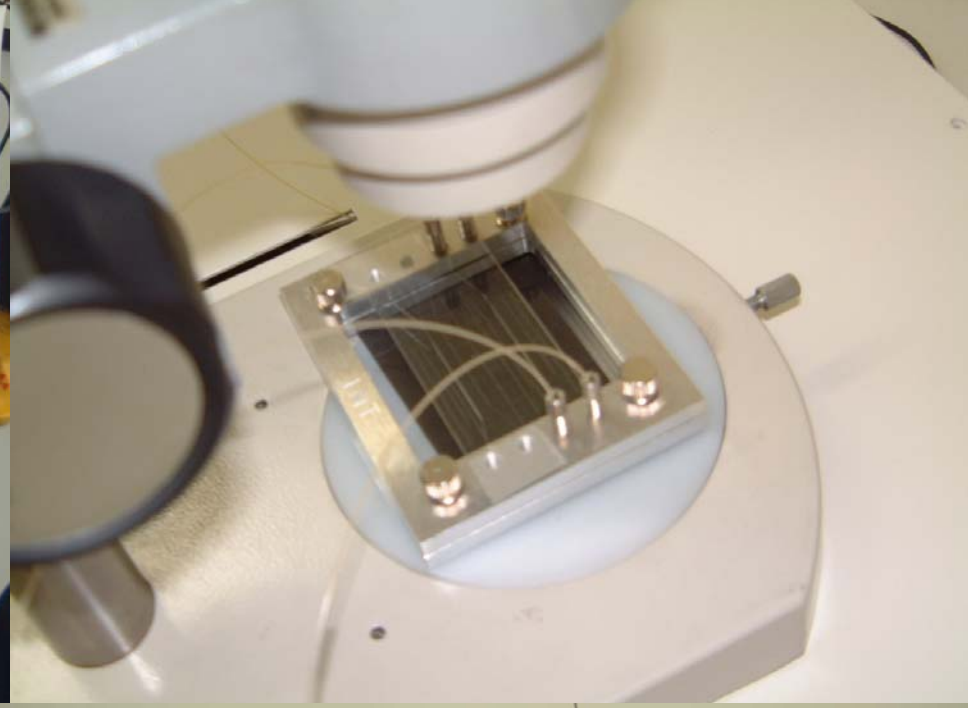


Fig. 3 Reaction conditions and results obtained with phase transfer diazocoupling reaction under microscale and macroscale conditions.

Microchip with Holder








マイクロチップ
気液二層流システム
V001
Gas
Flow Controller
V2 off
Flow Meter
V1 off
Pump
High
Pressure
Liquid
+383.66 µl
0.48 µl/min
16:40:58
ALOKA

Summary

- CANDLES III construction almost completed.
- The **preliminary** largest separation factor of Ca by LLE using DC18C6 is suggested**.This still needs to be checked** by TIMS, temp.& concentration dependence.
- We evaluated each contribution ratio of the field shift/hyperfine splitting shift effect to the mass effect of Ca for the 1st time.

  The contributon of the field shift effect is small, especially for ^{40}Ca-^{48}Ca, compared with Cr.
- These indications are promissing towards the mass production of enriched ^{48}Ca by the chemical separation method with the help of resins and/or microchannel chip.

Y. Fujii, Y. Sakuma, M. Tanimizu & A.Hibara(T. Kitamori)

Workshop on

Double Beta Decay and Isotope Science/Engineering

http://wwwkm.phys.sci.osaka-u.ac.jp/~hazama/iso-wsp/workshop.html

大阪大學

November 22(Tuesday) - 23(Wednesday), 2005 Osaka, Japan

Top Annoucement Program Workshop site Organizer Contact

- October 19: 1st announcement, update program pages
  November 22: Program and Participant List
  November 19: Workshop Poster
  November 19: Workshop Poster1, program

Motivation and scope

Neutrino mass is a key issue of current neutrino physics. Double beta decay may be the only probe presently able to access small neutrino masses with sensitivity down to ~0.03 eV, inferred from neutrino oscillation experiments. Actually, observation of neutrinoless double beta decay would identify a Majorana-type electron neutrino with a non-zero effective mass. Now the widest variety of stable isotopes are mainly produced at electromagnetic separators and gas centrifuges. Flexible highly efficient centrifugal technology is only possible for those elements (about 20) which have gaseous compounds at room temperature. Therefore, these methods cannot meet the production of some double beta decay isotopes such as ^{48}Ca, ^{96}Zr and ^{150}Nd etc. The workshop will provide wide ranging opportunities for scientists and engineers in various fields working not only on isotope effects in physics, chemistry, and engineering but also applications of isotopes in various fields in order to exchange new ideas, share current knowledge and present new results.

Topics:
Double beta decay
Isotope Science/Engineering
Mass Spectroscopy
Geochemistry, etc

Speakers:
R. Hazama, T. Kishimoto, I. Ogawa, A. Hibara,
N. Ishihara, K. Takeshita, Y. Murakami, M. Tanimizu,
K. Matsuoka, E. Matsumoto, Y. Fujii, K. Komura,
Y. Sakuma, S. Umehara, M. Miyabe, Y. Hirano
H. Niki, U. Tanaka,