Supported by Grant-in-Aid for Scientific Research (C) 24540295 and
Grant-in-Aid for Scientific Research on Innovative Areas 24104501

ジルコニウム96を用いたニュートリノを放出しない2重ベータ崩壊事象の探索

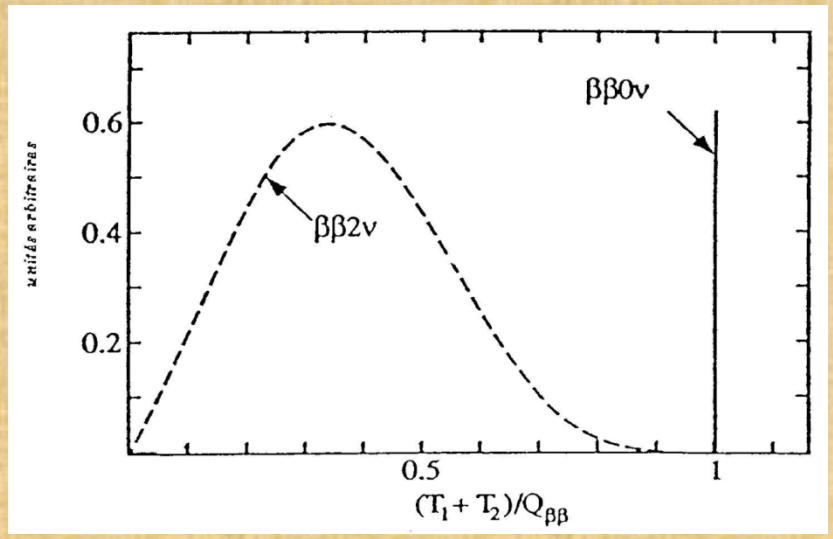
新学術領域「先端加速器LHCが切り拓くテラスケールの素粒子物理学」
研究会
2013年5月24日

宮城教育大学　教育学部
福田　善之

Neutrinoless double beta decay

$\beta\beta$ emitters with $Q_{\beta\beta}$ >2 Mev

Transition	$Q_{\beta\beta}$ (keV)	Abundance (%) ($^{232}Th = 100$)
$^{110}Pd \rightarrow ^{110}Cd$	2013	12
$^{76}Ge \rightarrow ^{76}Se$	2040	8
$^{124}Sn \rightarrow ^{124}Te$	2288	6
$^{136}Xe \rightarrow ^{136}Ba$	2479	9
$^{130}Te \rightarrow ^{130}Xe$	2533	34
$^{116}Cd \rightarrow ^{116}Sn$	2802	7
$^{82}Se \rightarrow ^{82}Kr$	2995	9
$^{100}Mo \rightarrow ^{100}Ru$	3034	10
$^{96}Zr \rightarrow ^{96}Mo$	3350	3
$^{150}Nd \rightarrow ^{150}Sm$	3667	6
$^{48}Ca \rightarrow ^{48}Ti$	4271	0.2



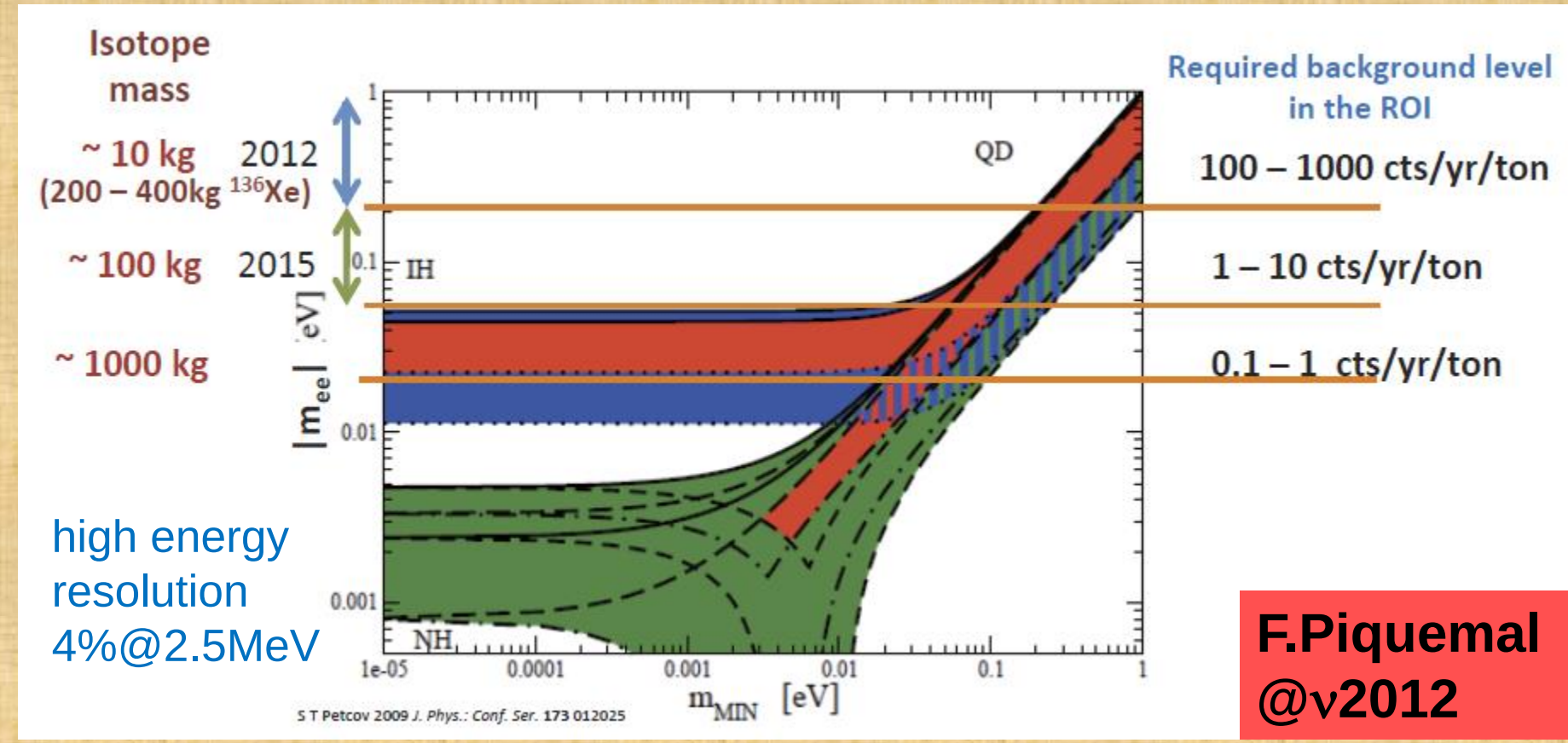
$$[T_{1/2}^{0\nu}(0^+ \rightarrow 0^+)]^{-1} = G_{0\nu}(E_0,Z)|M_{0\nu}|^2\langle m_\nu\rangle^2$$

$T_{1/2} \sim a(Mt/\Delta EB)$ a: abundance M: mass

t: meas.time ΔE: energy res. B: BG rate

Requirement : Low BG, Large target mass, High energy resolution

For future experiments

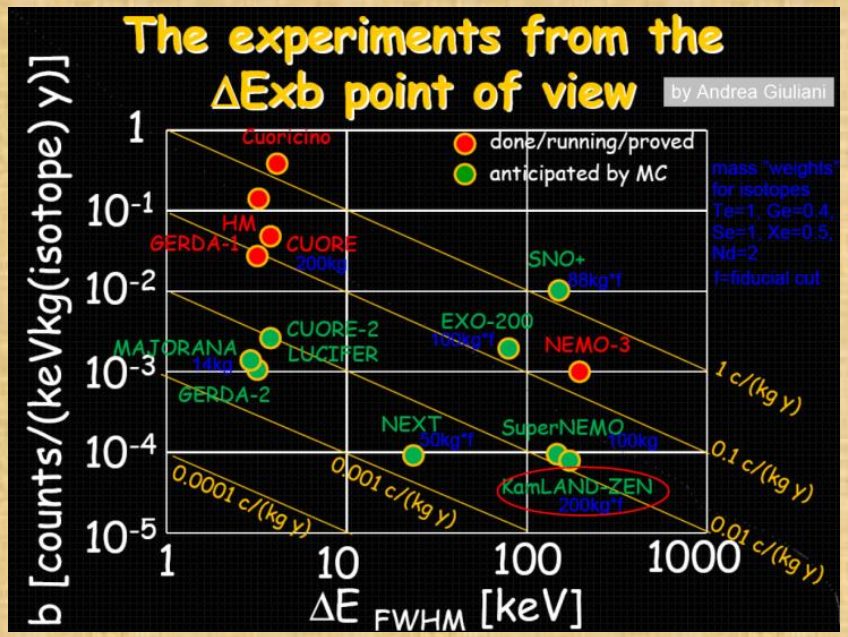


http://kds.kek.jp/getFile.py/access?contribId=37&sessionId=16&resId=2&materialId=slides&confId=9151

~tons of target will be needed for next generation detector

Future experiments for Neutrinoless double beta decay

- Experimental requirement for BG rate and ΔE

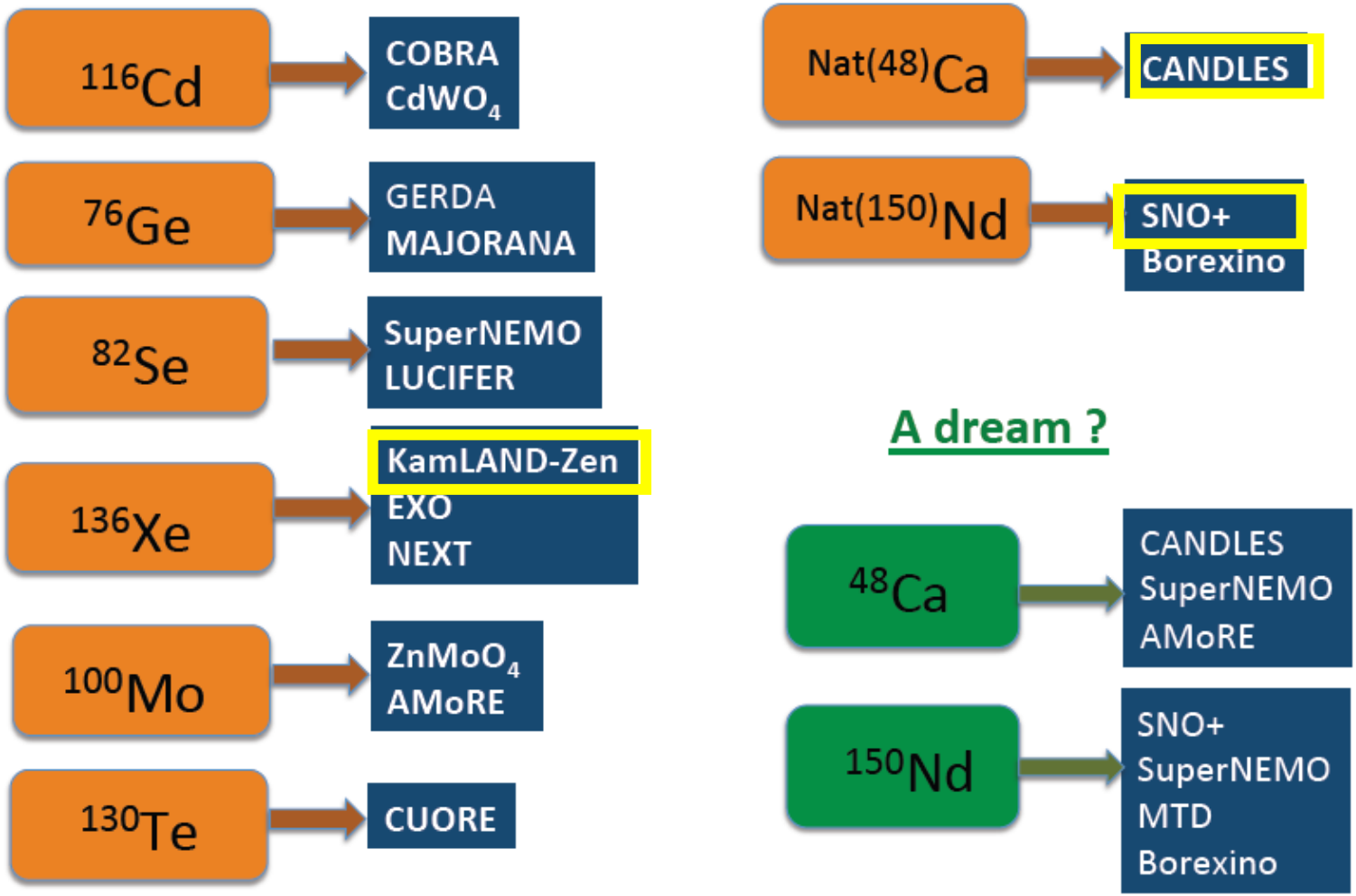


- To achive m_ν < 100meV
  - high energy resolution 4%@2.5MeV
  - low background rate 0.01count kg^{-1} y^{-1}
  - ton scale of target

Liq. Scintillator is easy to scale up target volume

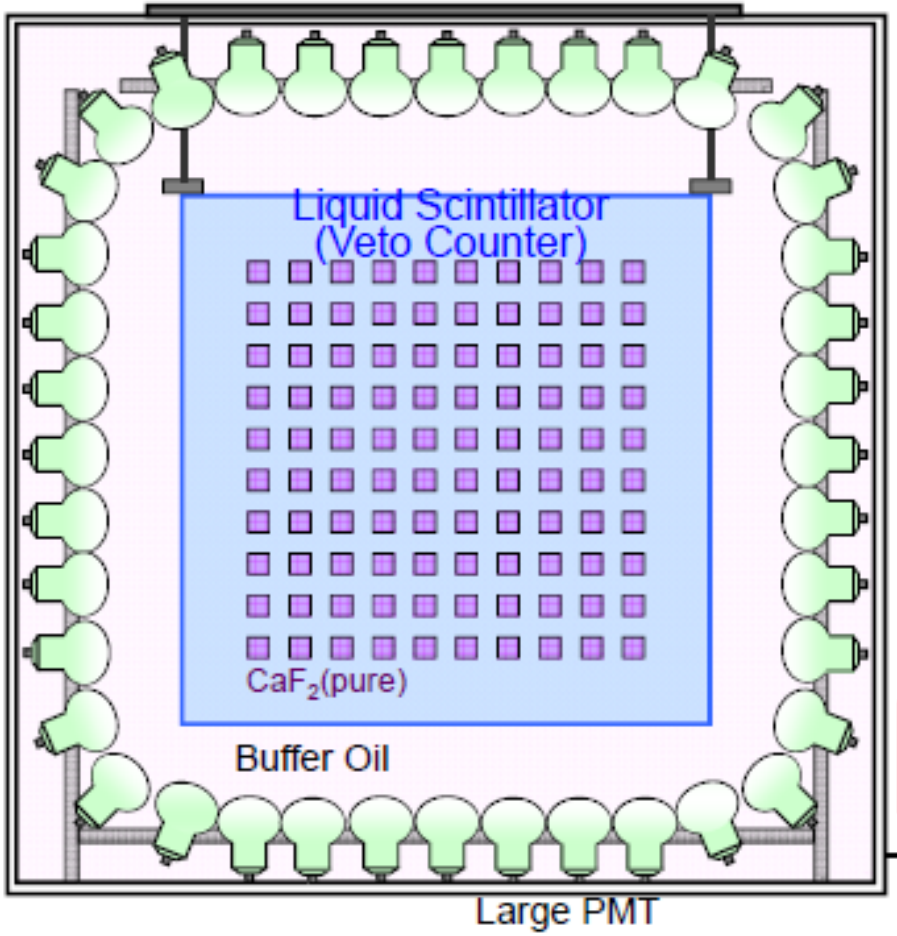
Studied isotopes

Piquemal@ν2012



Design Concepts of CANDLES

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



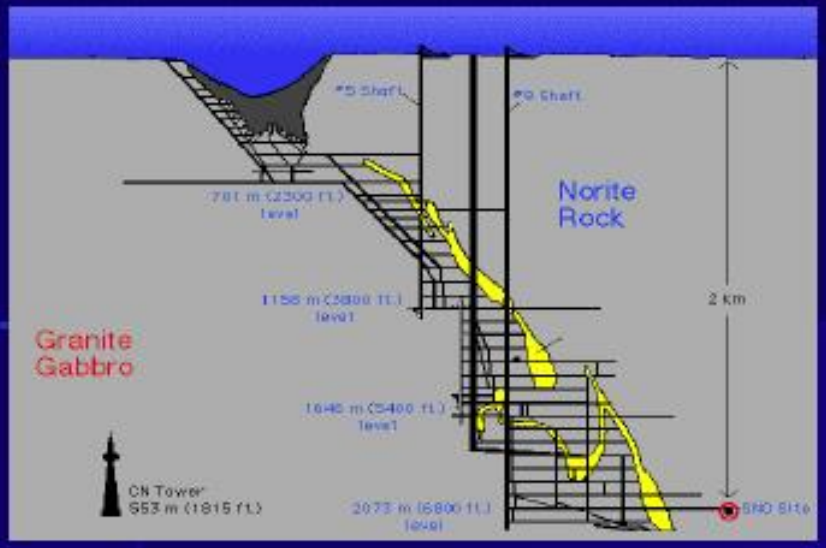
- CaF_2(pure) scintillator
  - Long attenuation length (>10m@350nm)
  - Double beta decay source
    ^{48}Ca (Q_{bb}=4.27MeV)
- Liquid scintillator
  - 4 π Active Shield
- Large photomultiplier tube
  - Signals from both scintillators are detected simultaneously

⇩

- Active Shielding Technique

Different time constants
CaF_2(pure) : ~1μsec
Liquid scintillator : a few 10 nsec

UMEHARA Saori, 16th Nov. 2011, DBD11



SNO+

~~1000 tonnes D_2O~~ → **780 tonnes liquid Scintillator (LAB)**

12 m diameter Acrylic Vessel

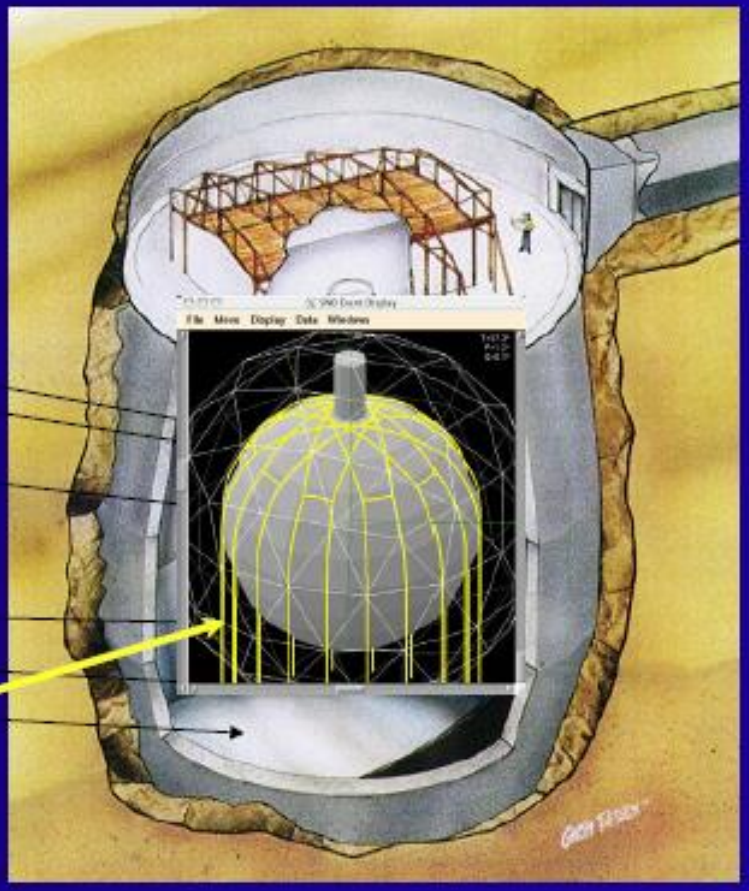
18 m diameter support structure; 9500 PMTs

(~60% photocathode coverage)

1700 tonnes inner shielding H_2O
5300 tonnes outer shielding H_2O
Urylon liner radon seal

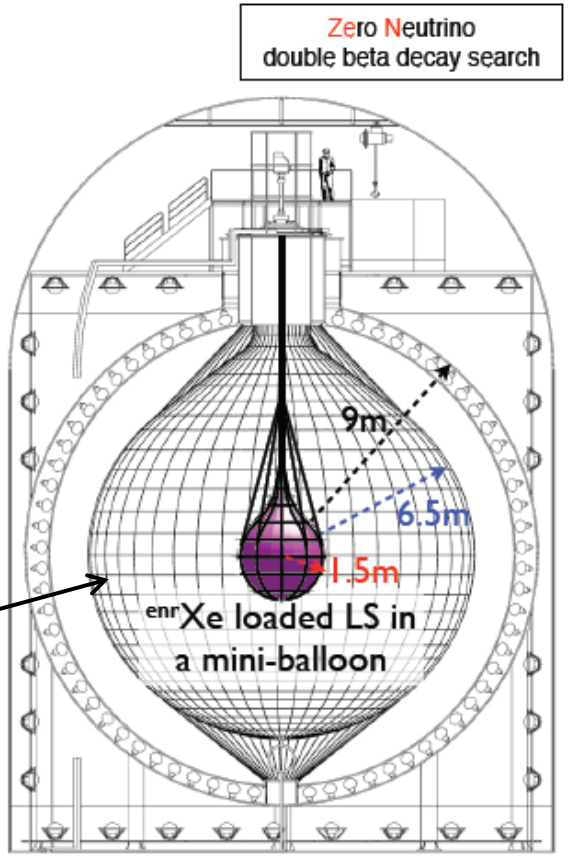
hold down rope net

depth: 2092 m (~6010 m.w.e.) ~70 muons/day



KamLAND-Zen

K.Inoue@ν2012



idea to load Xe into LS is from Raju PRL72,1411(1994)

~320kg 90% enriched ^{136}Xe installed so far
total 600+ kg in the mine
production reaches 700kg in this year

Pseudo-cumene 20%
+
Dodecane 80%
+
PPO 0.15wt%

Good features of us

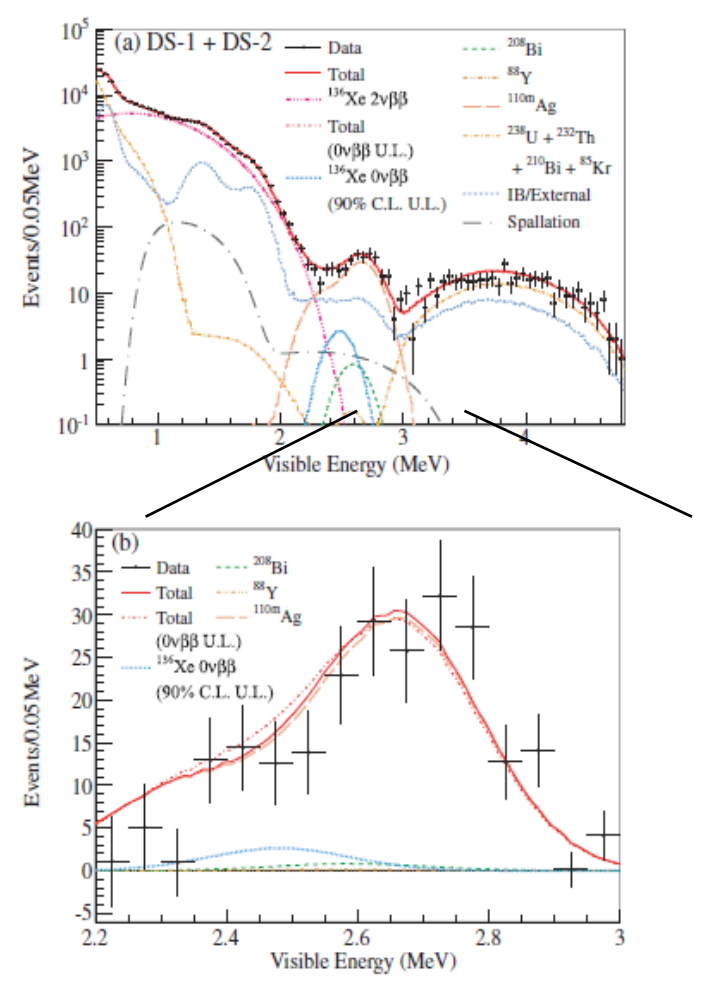
- running detector
  → relatively low cost and quick start
- huge and clean (1200m^3, U: 3.5x10^{-18} g/g, Th: 5.2x10^{-17})
  → negligible external gamma
  (Xe and mini-balloon need to be clean)
- Xe-LS can be purified, mini-balloon replaceable if necessary, with relatively low cost
  → highly scalable (up to several tons of Xe)
- No escape or invisible energy from β, γ
  → BG identification relatively easy
- anti-neutrino observation continues
  → geo-neutrino w/o japanese reactors

Disadvantages toward an ultimate sensitivity

× relatively poor energy resolution
tolerable thanks to slow 2ν2β and low BG

× no β/γ discrimination so far
× delicate balloon film
× limited LS composition (for density matching)

Energy spectrum obtained by KamLAND



- Most possible BG
  - ^{110m}Ag (β-γ, β_{max}=3,02MeV, τ=360d)
  - ^{214}Bi on IB (β-γ, β_{max}=3,27MeV, τ=15d)
  - Half-life of 0νββ

$T^0_{1/2} > 1.9 \times 10^{25}$ y(90%C.L.)

  - neutrino mass limit (conbined by EXO200)

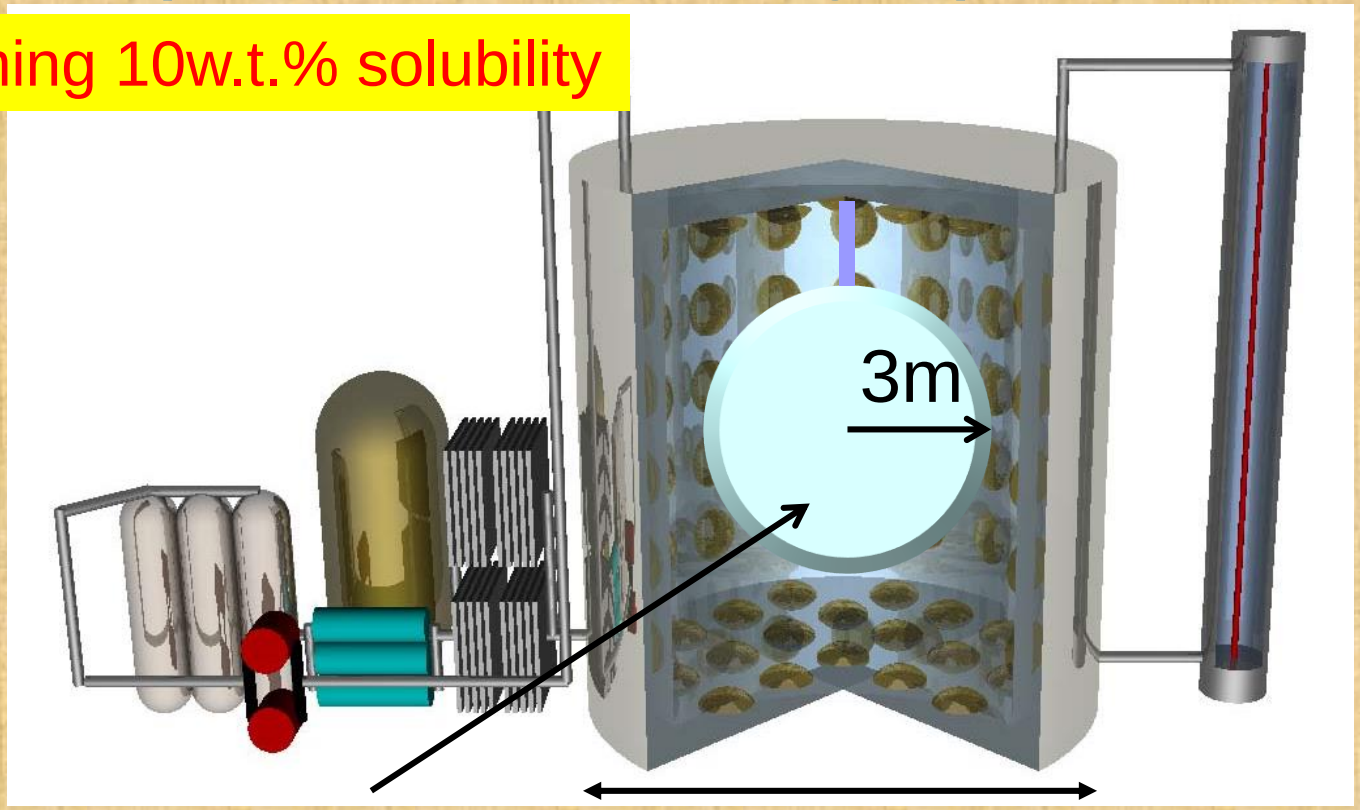
$m\nu < 120$ -250mev

PRL 110,062502 (2013)

Detector design for ZICOS experiment

- **Zirconium Complex in Organic liquid Scintillator (ZICOS) for double beta decay experiment**

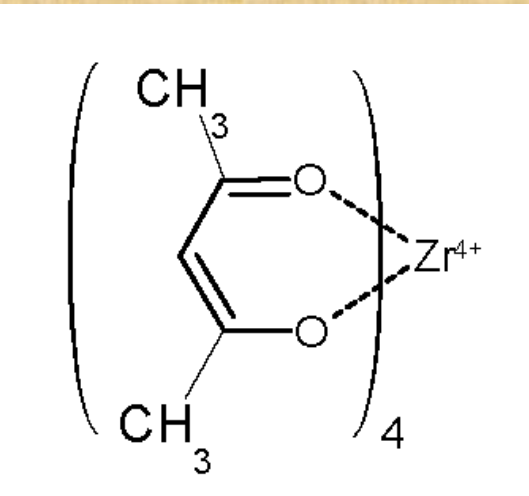
Assuming 10w.t.% solubility



Zirconium loaded 100ton LS

Zirconium β-diketon complex

- Zirconium(IV) acetylacetonate ($Zr(acac)_4$)

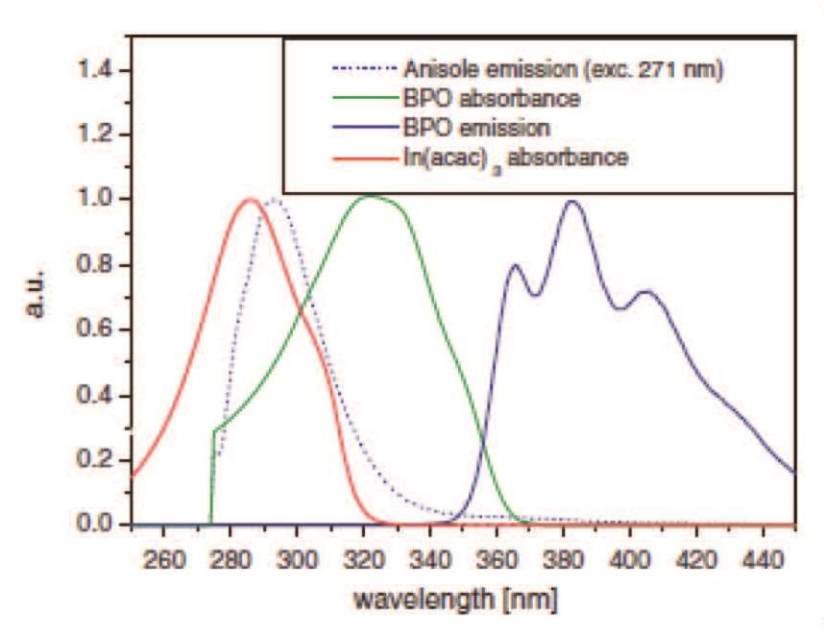


Molecular weight : 487.66

- Advantage
  - good solubility (over 10w.t.%) in Anisole (PhOMe)
  - Stable and cheep
  - Commercial product
- Disadvantage
  - Low scintillation light yield

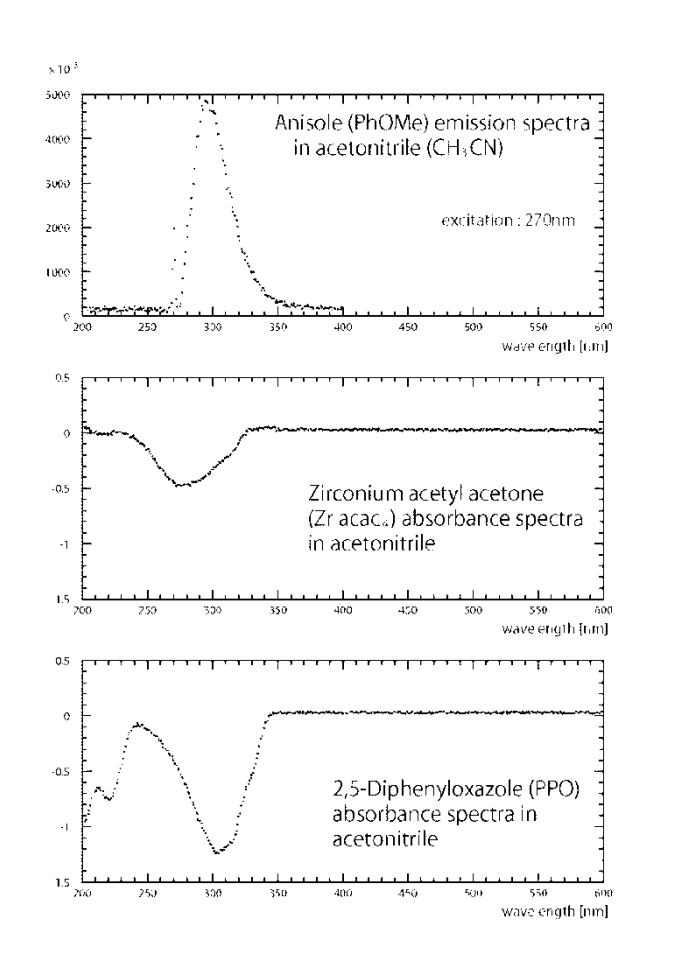
What's problem

- Absorption spectra of $In(acac)_3$ (indium acetyl acetone) was overlapped with the emission spectra from Anisole（Chem. Phys. Lett., 435(2007), 252)



Same overlap between the emission and the absorption could be occurred even if different metal (Zr) was used.

Observed absorption spectra of $Zr(acac)_4$



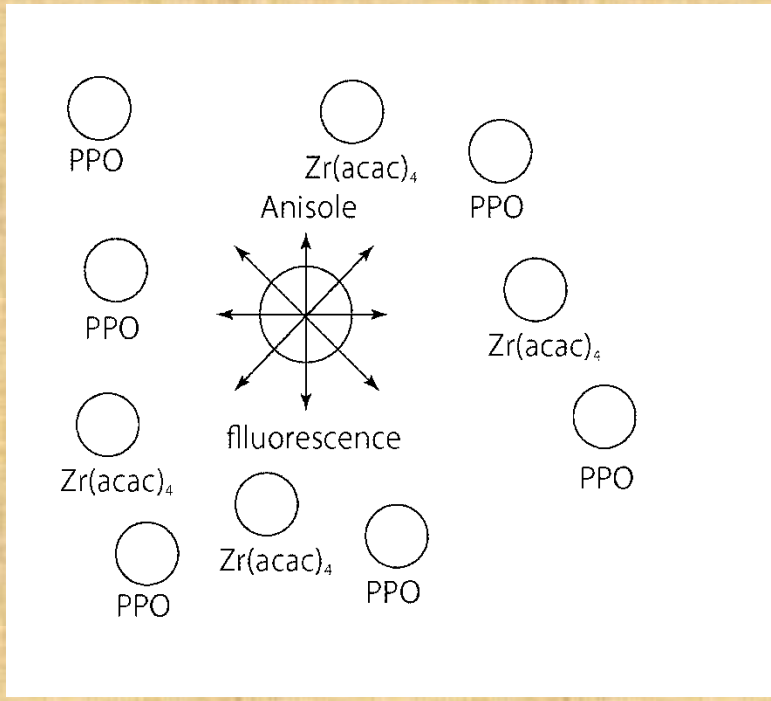
- Emission peak of anisole was observed around 295nm.
- Absorption peak of $Zr(acac)_4$ was observed around 270nm.

Scintillation light from anisole (PhOMe) might be absorbed by $Zr(acac)_4$

Simple expectation for quenching

- Assuming to same cross section for light

$$\text{Light yield} = L_0 \times \frac{\sigma_1 N_{ppo}}{\sigma_1 N_{ppo} + \sigma_2 N_{Zr}}$$

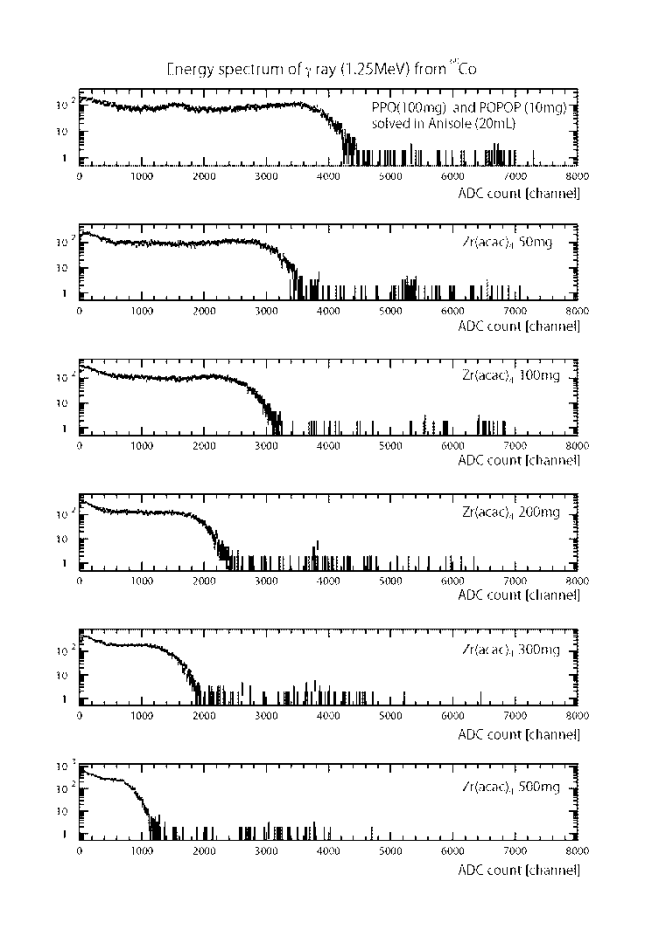


L_0 : Light yield of anisole + PPO + POPOP

N_{ppo} , N_{Zr} : Number of molecular for PPO and $Zr(acac)_4$

σ_1, σ_2 : cross section of absorbance for PPO and $Zr(acac)_4$

Scintillation Light yield (^{60}Co) with respect to concentration of $Zr(acac)_4$

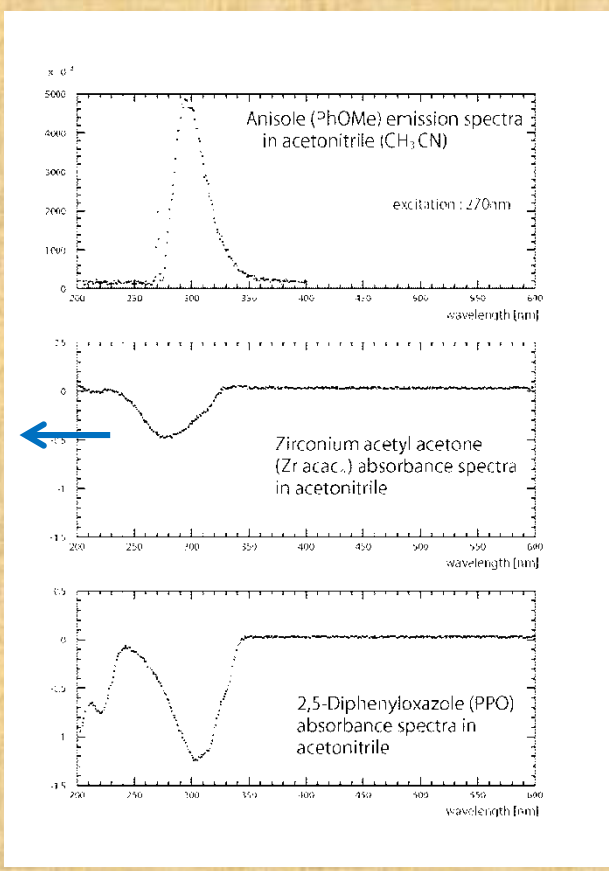


concentration of $Zr(acac)_4$	Observed channel	Expected channel
0 mg	3850	3850
50mg ($1.03X10^{-4}$)	3175	3138
100mg ($2.05X10^{-4}$)	2800	2651
200mg ($4.10X10^{-4}$)	2000	2018
300mg ($6.15X10^{-4}$)	1600	1613
500mg ($1.03X10^{-3}$)	900	1178

PPO 100mg : $4.52X10^{-4}$ mol

Improvement of scintillation light yield

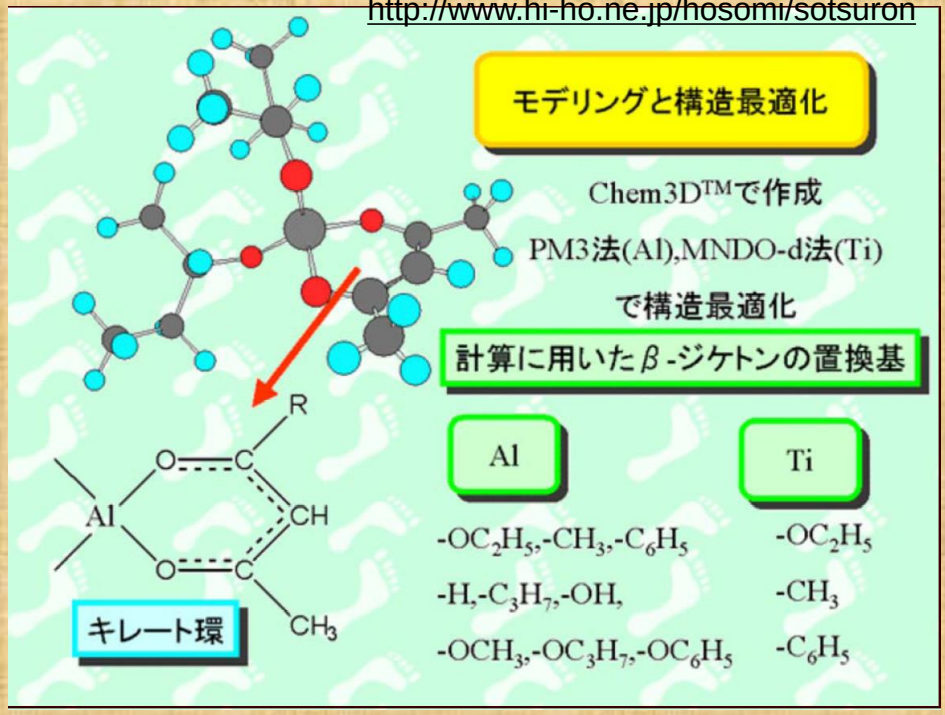
- Move absorption peak to shorter wavelength



- How to do it?

substituent groups

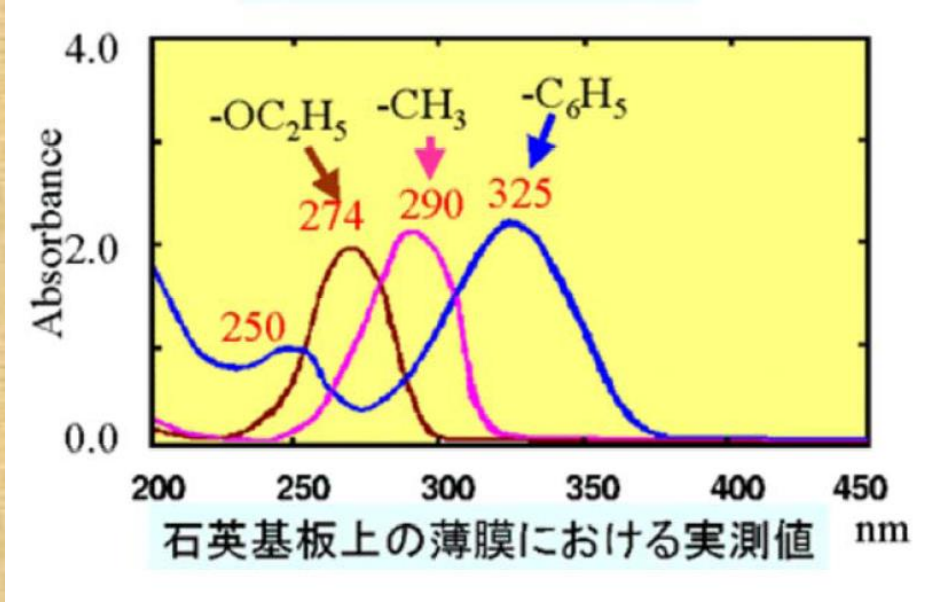
http://www.hi-ho.ne.jp/hosomi/sotsuron



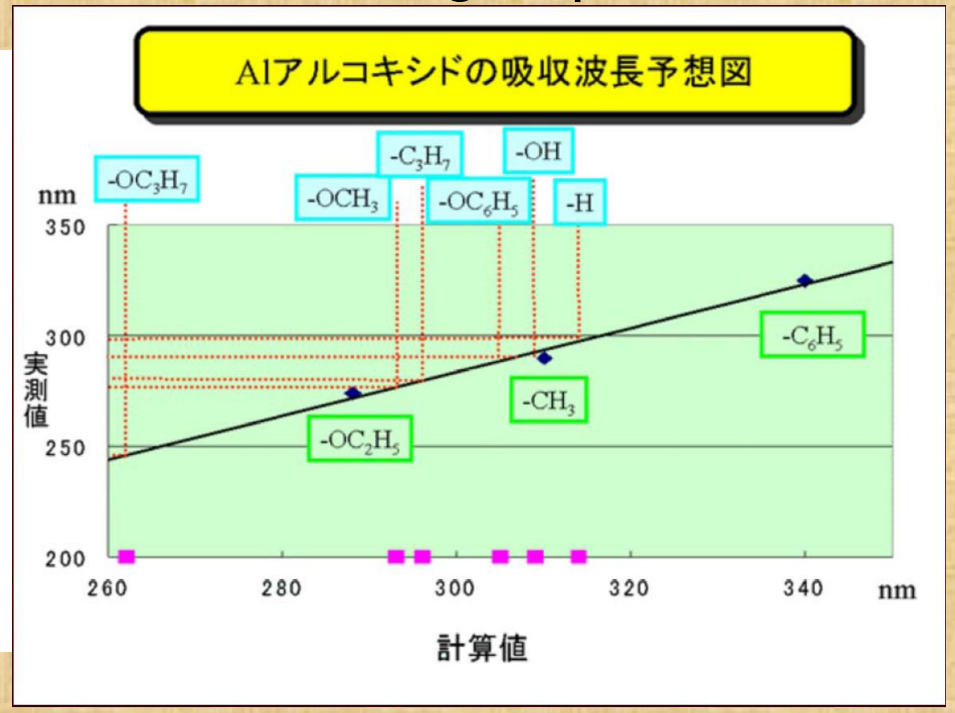
Courtesy of Prof. Yoshiyuki Kowada (Hyogo University of Education)

Absorbance peak for several substituent groups

- Measured absorbance peaks for several substituent groups

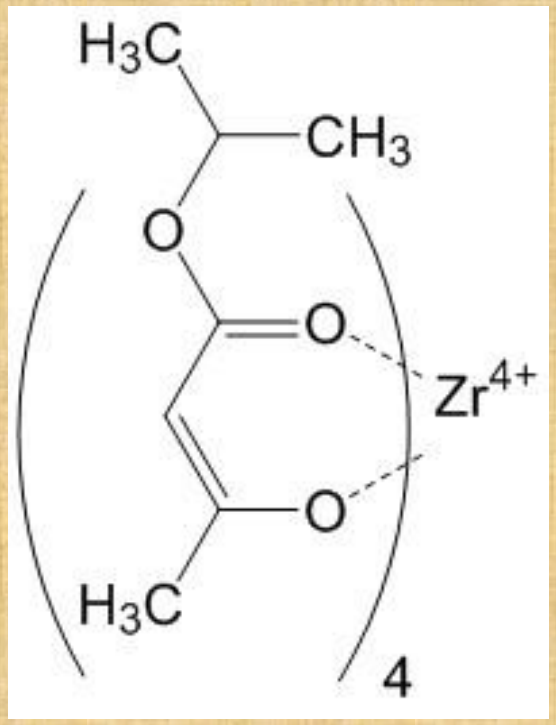


- Expected absorbance peak for several substituent groups

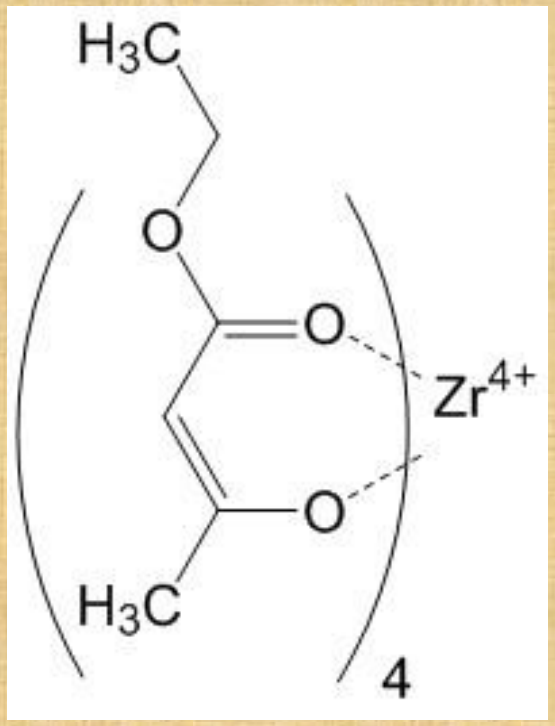


Zr β-diketon complex introducing substituent groups (β-keto ester complex)

Zr(CH3COCHCOOCH(CH3))$_4$ = Zr(iprac)$_4$
mw : 711.92



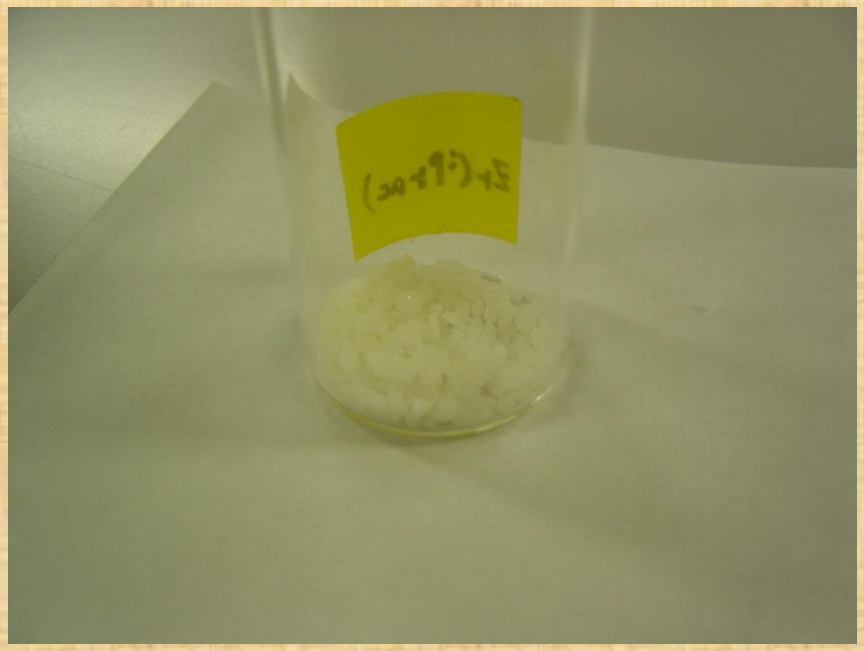
Zr(CH3CCOCHCOOCH3)$_4$ = Zr(etac)$_4$
mw : 665.81



Zr β-keto ester complex

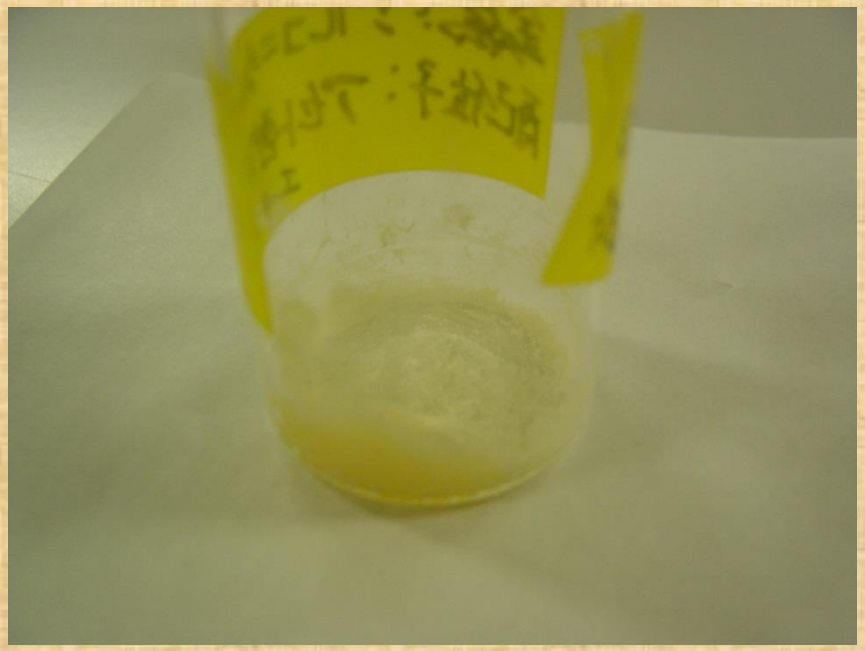
$Zr(iprac)_4+(iprac)_{1.5}$

state: powder



$Zr(etac)_4$

state : dry solid

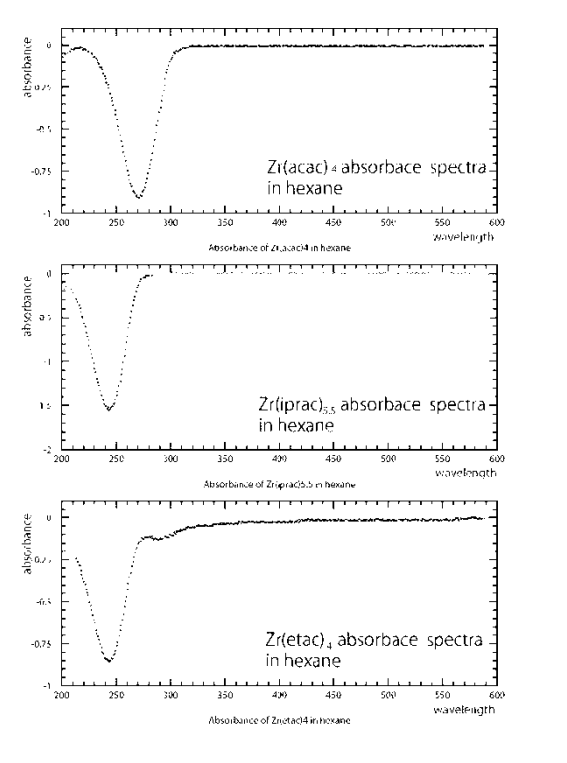


Synthesized by Prof. Takahiro Gunji (Tokyo University of Science)

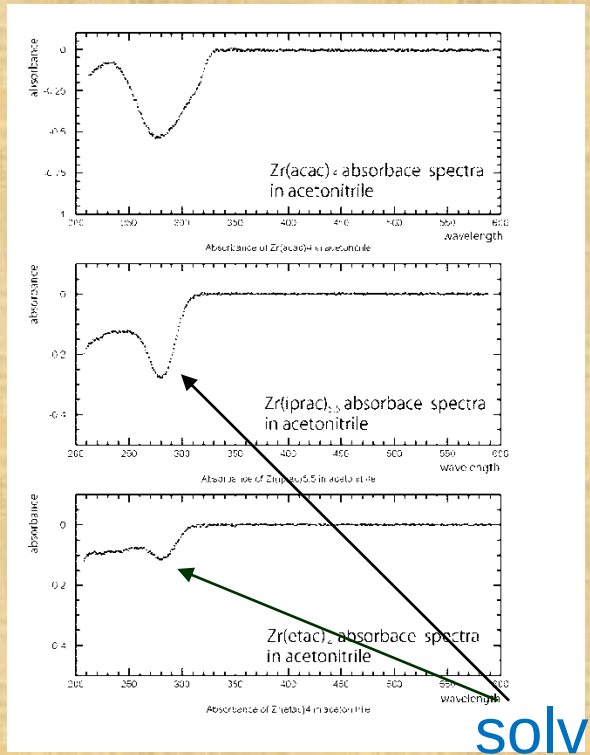
Solubility > 10 w.t.% for anisole

Absorbance spectra (Solvent effect)

Solution : Hexane



Solution : acetonitrile

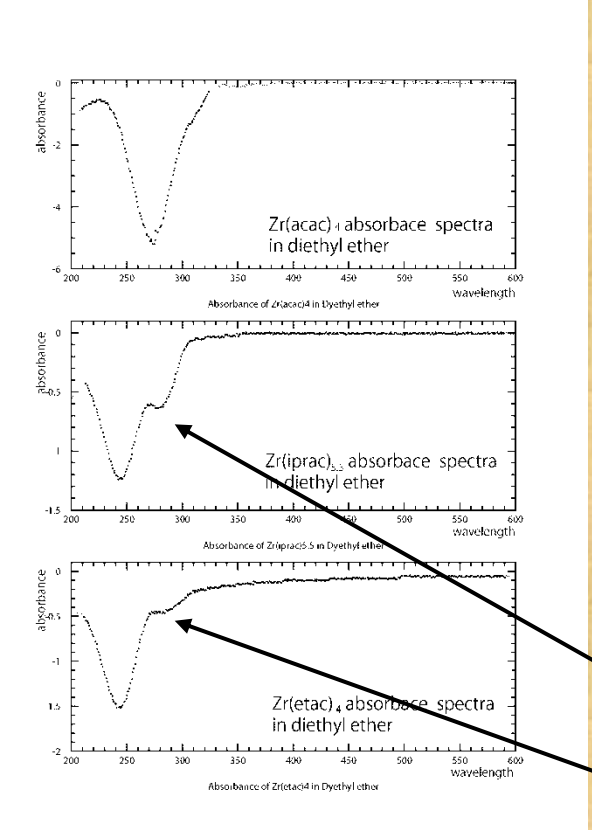


solvent effect

Absorption peak moves to shorter wavelength, however it depends on the polarity of solvent.

Absorbance in liquid scintillator

Solution : Diethyl Ether

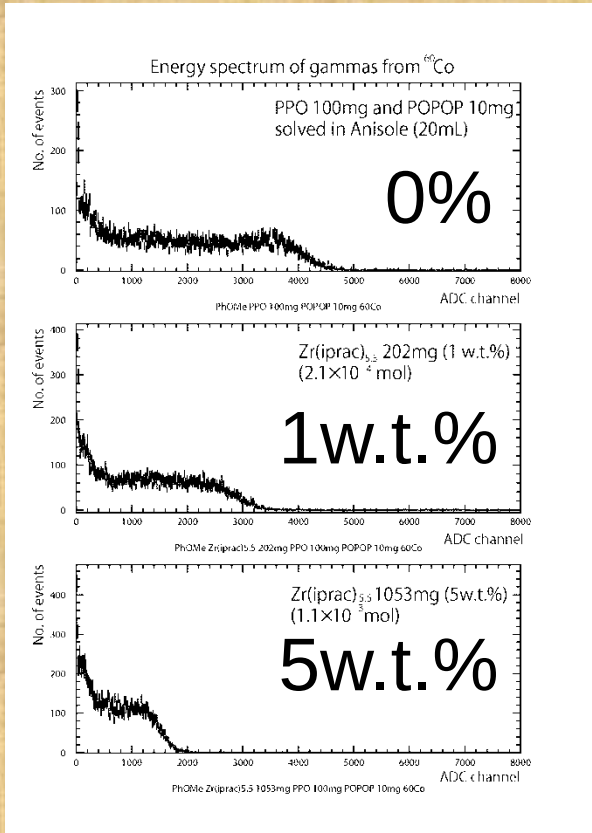


- Solvent effect could depend on the polarity (dielectric const.)
  - Acetonitrile : 37.5
  - Hexane : 1.89
  - Anisole : 4.3
- Need solution which has same polarity as anisole
  - Diethyl ether : 4.33

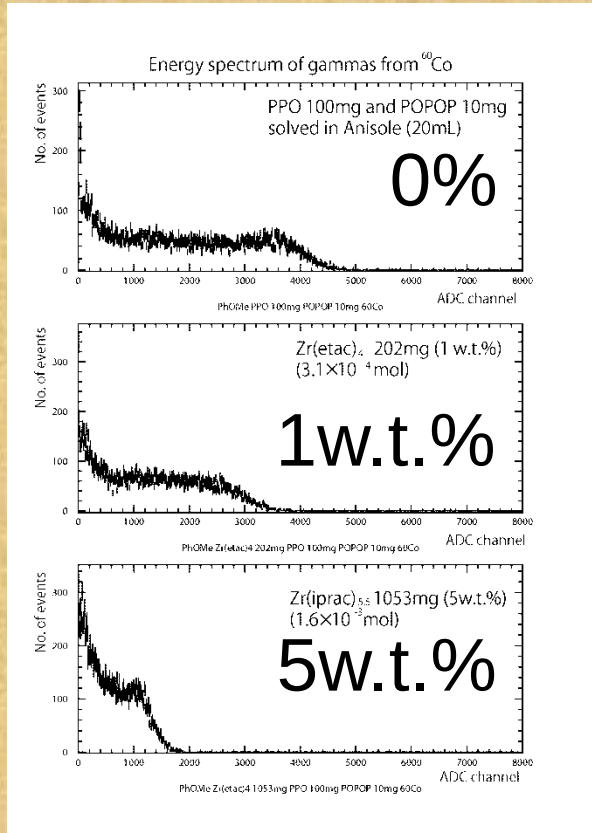
Still solvent effect remains around 270nm

Light yield of Zr β-keto ester scintillator

$Zr(iprac)_{5.5}$ in anisole



$Zr(etac)_4$ in anisole



Same quenching as $Zr(acac)_4$ was observed

Requirement of scintillator solvent

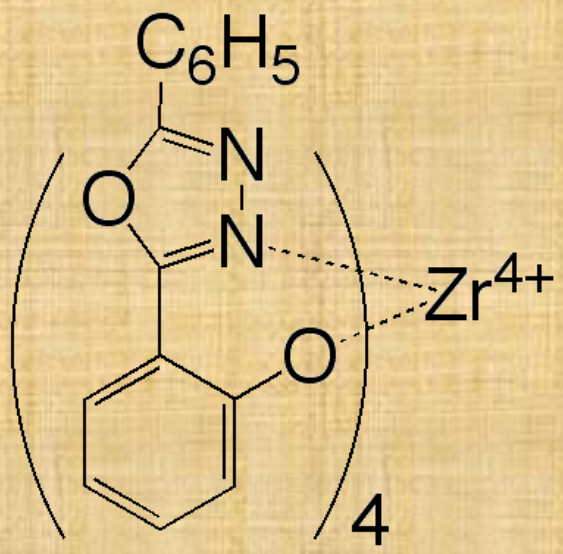
- Low polarity (low dielectric constant)
  - No absorption ~270nm
- Aromatic compounds
  - luminescence >270nm
- Keep high solubility > ~10%
- Keep luminescence from solvent

Possible solvent:
Toluene / Xylene

Solvent	mp	bp	D_4^{20}	n_D^{20}	ε	R_D	μ
Acetic acid	17	118	1.049	1.3716	6.15	12.9	1.68
Acetone	-95	56	0.788	1.3587	20.7	16.2	2.85
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Anisole	-3	154	0.994	1.517	4.33	33	1.38
Benzene	5	80	0.879	1.5011	2.27	26.2	0
Bromobenzene	-31	156	1.495	1.558	5.17	33.7	1.55
Carbon disulfide	-112	46	1.274	1.6295	2.6	21.3	0
Carbon tetrachloride	-23	77	1.594	1.4601	2.24	25.8	0
Chlorobenzene	-46	132	1.106	1.5248	5.62	31.2	1.54
Chloroform	-64	61	1.489	1.4458	4.81	21	1.15
Cyclohexane	6	81	0.778	1.4262	2.02	27.7	0
Dibutyl ether	-98	142	0.769	1.3992	3.1	40.8	1.18
o -Dichlorobenzene	-17	181	1.306	1.5514	9.93	35.9	2.27
1,2-Dichloroethane	-36	84	1.253	1.4448	10.36	21	1.86
Dichloromethane	-95	40	1.326	1.4241	8.93	16	1.55
Diethylamine	-50	56	0.707	1.3864	3.6	24.3	0.92
Diethyl ether	-117	35	0.713	1.3524	4.33	22.1	1.3
1,2-Dimethoxyethane	-68	85	0.863	1.3796	7.2	24.1	1.71
N,N -Dimethylacetamide	-20	166	0.937	1.4384	37.8	24.2	3.72
N,N -Dimethylformamide	-60	152	0.945	1.4305	36.7	19.9	3.86
Dimethyl sulfoxide	19	189	1.096	1.4783	46.7	20.1	3.9
1,4-Dioxane	12	101	1.034	1.4224	2.25	21.6	0.45
Ethanol	-114	78	0.789	1.3614	24.5	12.8	1.69
Ethyl acetate	-84	77	0.901	1.3724	6.02	22.3	1.88
Ethyl benzoate	-35	213	1.05	1.5052	6.02	42.5	2
Formamide	3	211	1.133	1.4475	111	10.6	3.37
Hexamethylphosphoramide	7	235	1.027	1.4588	30	47.7	5.54
Isopropyl alcohol	-90	82	0.786	1.3772	17.9	17.5	1.66
Methanol	-98	65	0.791	1.3284	32.7	8.2	1.7
2-Methyl-2-propanol	26	82	0.786	1.3877	10.9	22.2	1.66
Nitrobenzene	6	211	1.204	1.5562	34.82	32.7	4.02
Nitromethane	-28	101	1.137	1.3817	35.87	12.5	3.54
Pyridine	-42	115	0.983	1.5102	12.4	24.1	2.37
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Toluene	-95	111	0.867	1.4969	2.38	31.1	0.43
Trichloroethylene	-86	87	1.465	1.4767	3.4	25.5	0.81
Triethylamine	-115	90	0.726	1.401	2.42	33.1	0.87
Trifluoroacetic acid	-15	72	1.489	1.285	8.55	13.7	2.26
2,2,2-Trifluoroethanol	-44	77	1.384	1.291	8.55	12.4	2.52
Water	0	100	0.998	1.333	80.1	3.7	1.82
[illegible] -Xylene	-25	144	0.88	1.5054	2.57	35.8	0.62

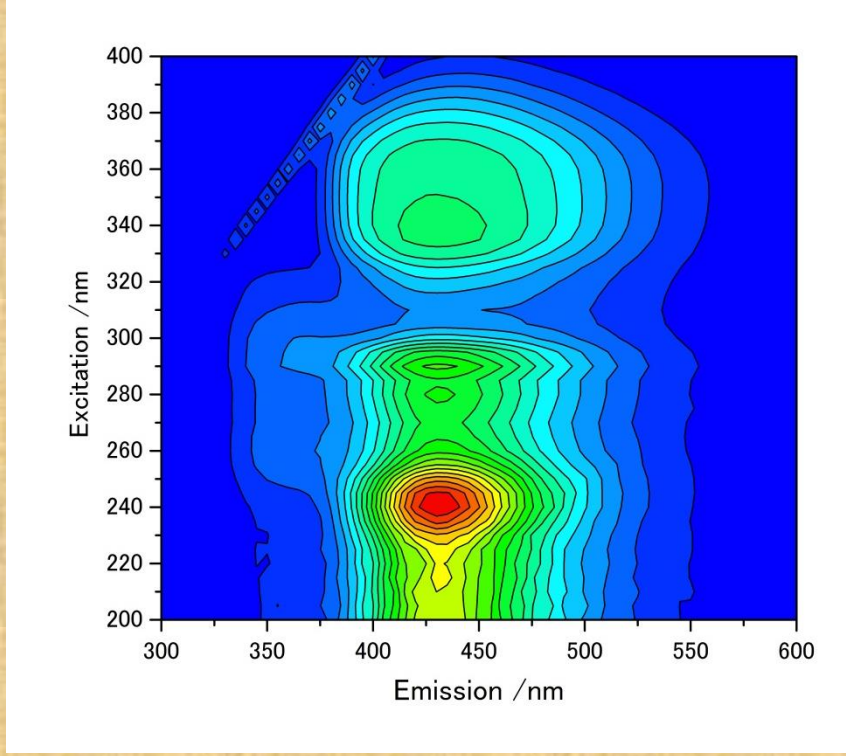
Zirconium complex with luminescence

- Zr-ODZ complex



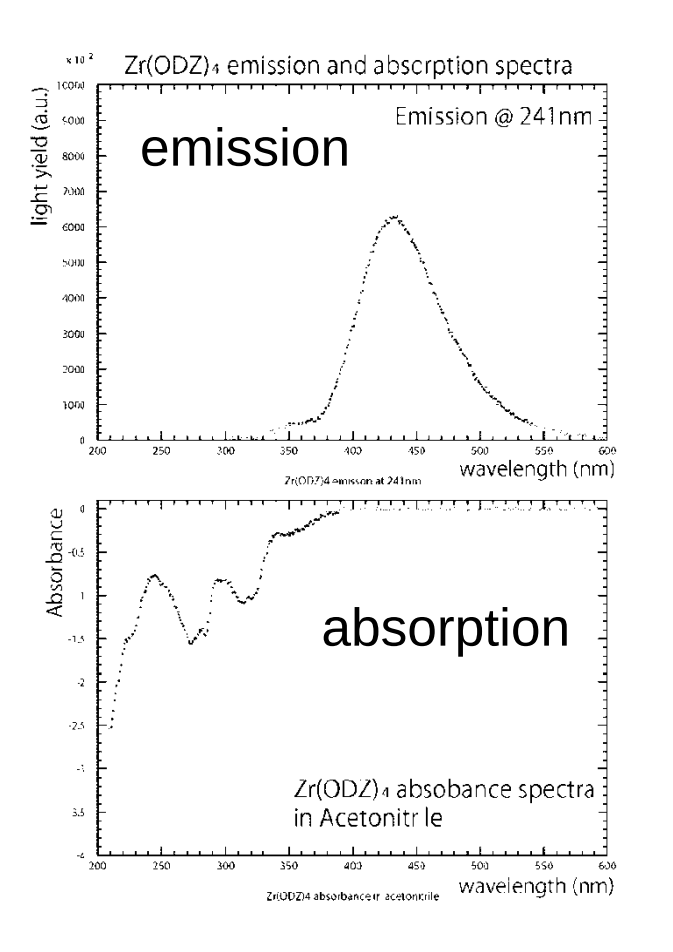
m.w. = 1040.18

- Photo luminescence



- **Solvent : Acetonitrile**
- **Concentration : 3.0×10^{-5} mol/L**

Emission and absorption spectrum of $Zr(ODZ)_4$



- Emission wavelength : 430nm

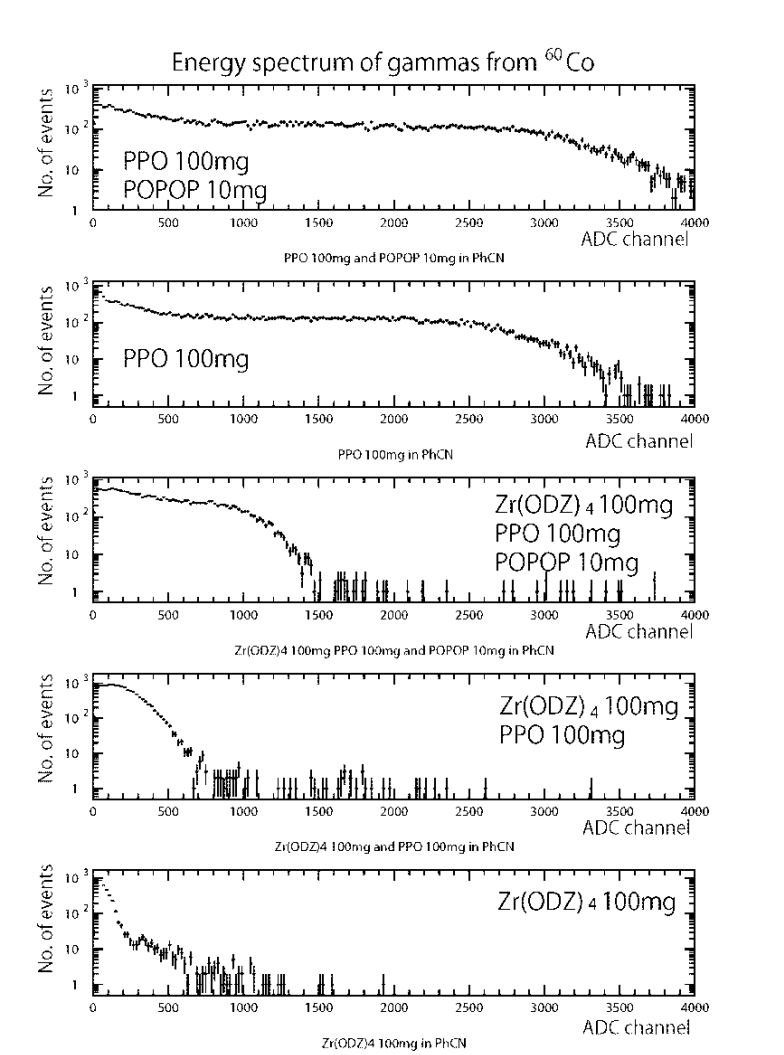
PMT sensitive

- Absorption wavelength: 270nm and 320nm

different from excitation W.L.

- Solvent : PhCN (Benzonitrile)
- Solubility : ~5w.t.%

Response for γ-irradiation



- Most of emission light from PhCN was not used for the emission of $Zr(ODZ)_4$.
- third excitation of ~340nm from PPO was used for the emission of $Zr(ODZ)_4$.
- Estimated Quantum yield was obtained ~30% at first excitation of ~240nm.

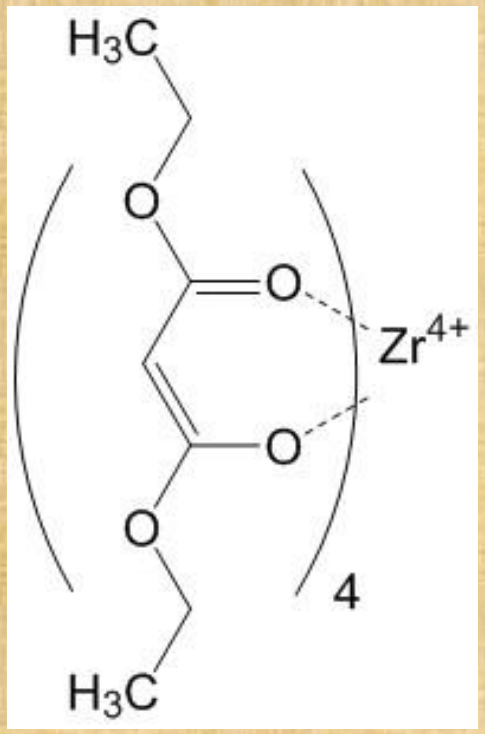
Need another solvent which has shorter emission wavelength than PhCN.

Summary

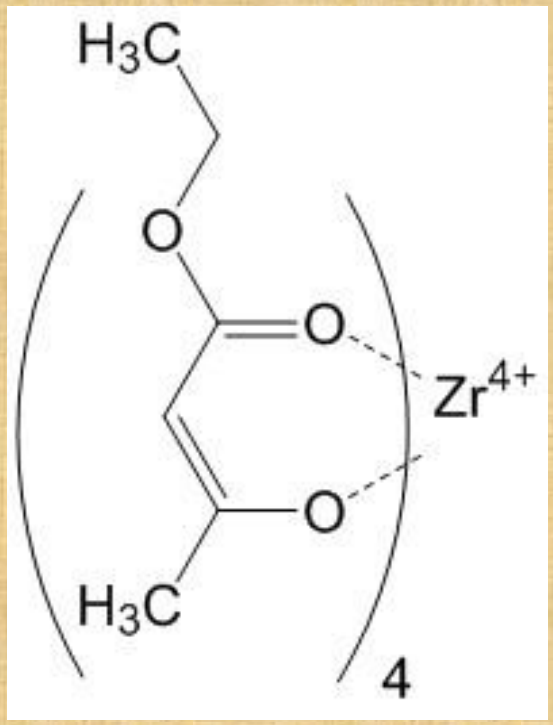
- High solubility of Zirconium β-keto ester in Anisole (>~10w.t.%) was achieved.
- Confirmed absorption peak moves to shorter wavelength (275nm → 245nm) by introducing ester substituent groups.
- Observed scintillation light yield decreased in proportion to the concentration of Zr β-keto ester complex due to solvent effect → Need low polarity solvent
- To avoid solvent effect, we will synthesize new complex tetrakis (diethyl malonato) zirconium.

Tetrakis (diethyl malonato) Zr compex

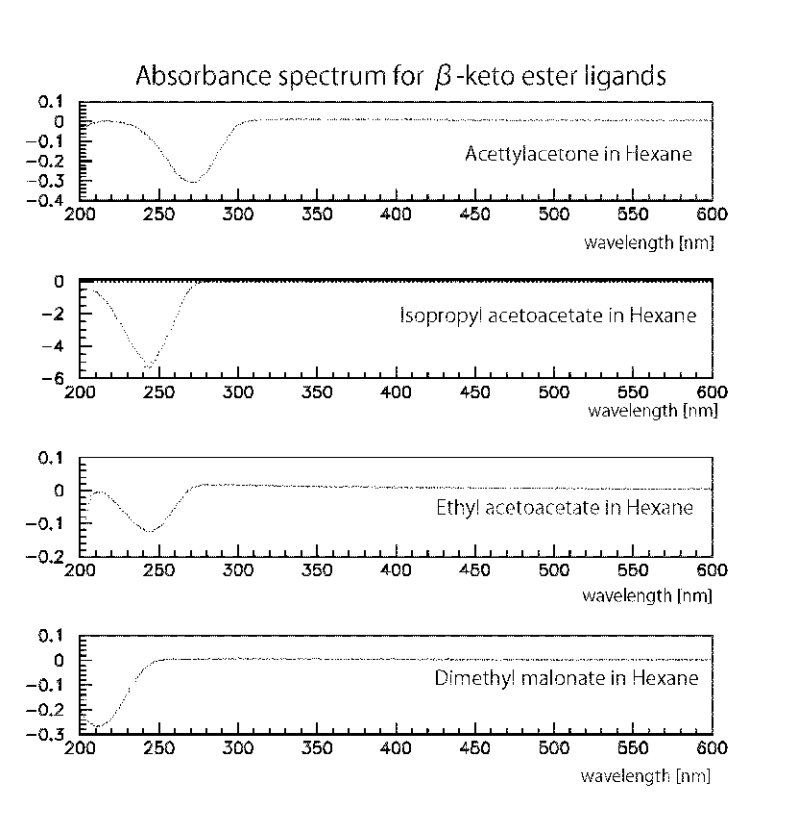
$Zr(CH_3CH_2OCOCHCOOCH_2CH_3)_4$ = $Zr(deml)_4$
mw : 727.84

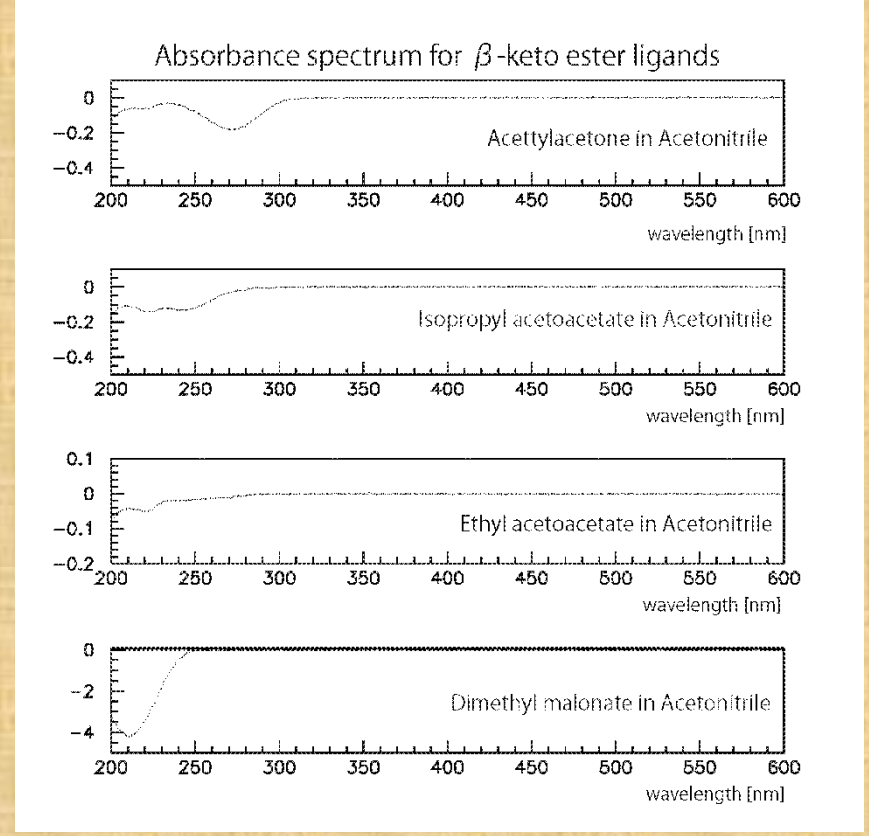


$Zr(CH_3CCOCHCOOCH_3)_4$ = $Zr(etac)_4$
mw : 665.81



Absorbance spectrum for β-keto ester ligands

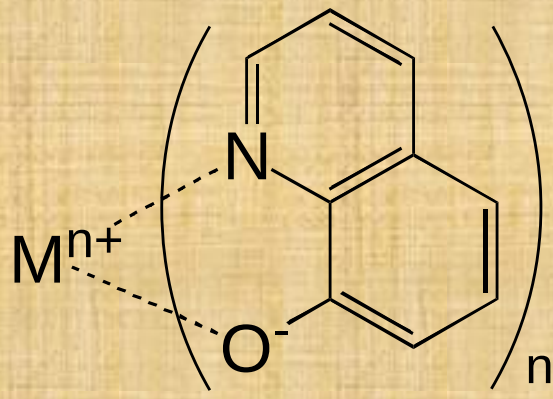




BACKUP

Tetrakis 8-quinolinolate Zr complex loaded scintillator

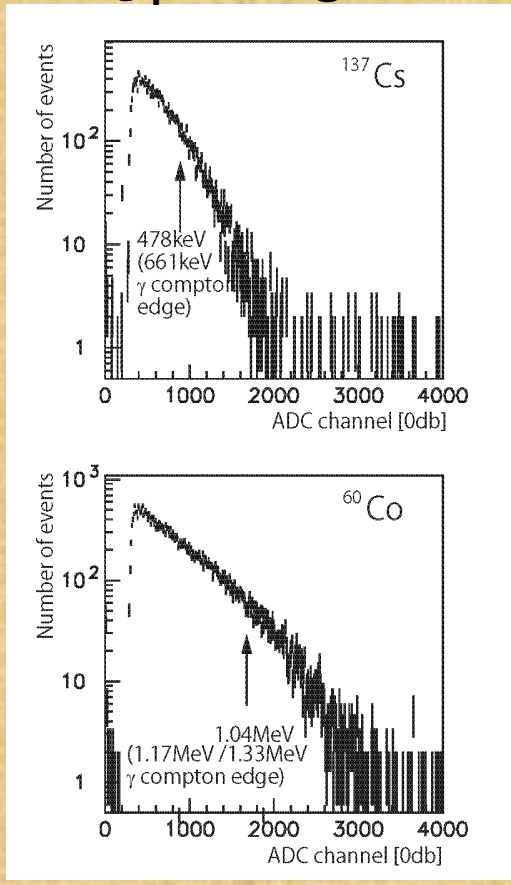
- Tetrakis (8-quinolinolate) Zirconium complex (ZrQ_4)



M = In, n = 3; M = Zr, n = 4

ZrQ_4 m.w. = 689.07

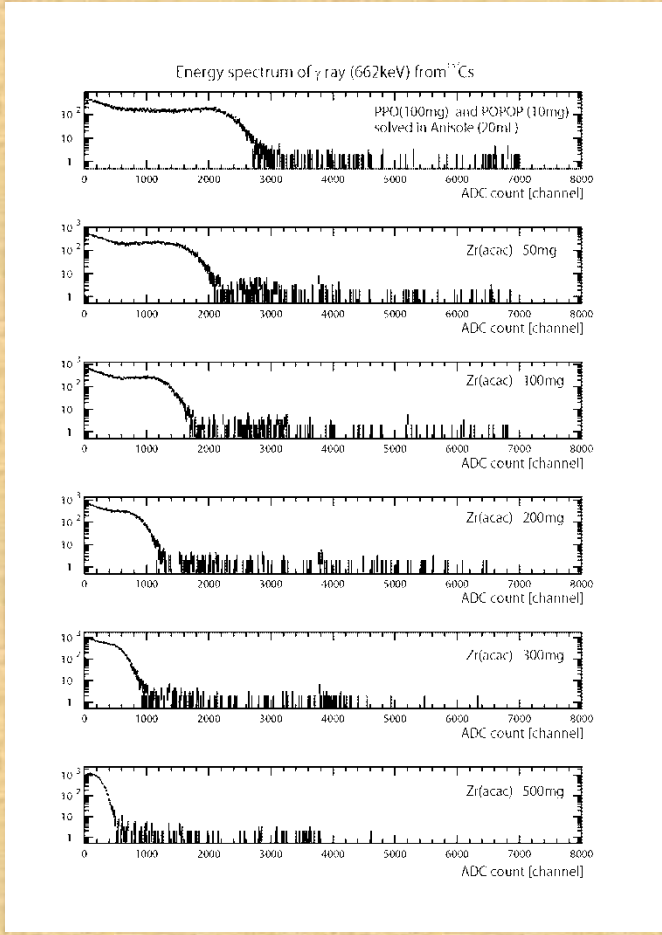
ZrQ_4 50mg in PhCN-POPOP



Quantum Yield=1.1% obtained by optical method

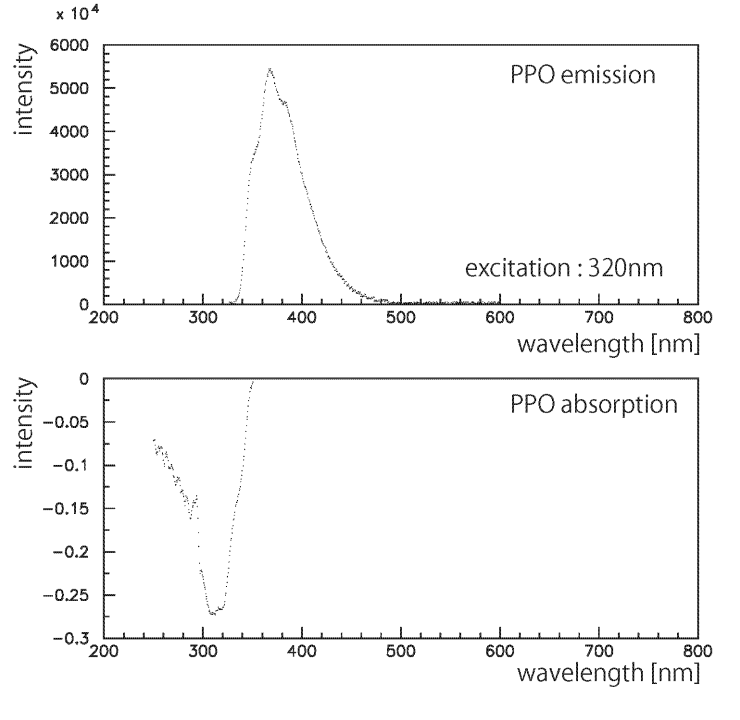
Light Yield to BC505: =7.3%

Scintillation light yield (^{137}Cs) with respect to concentration of $Zr(acac)_4$



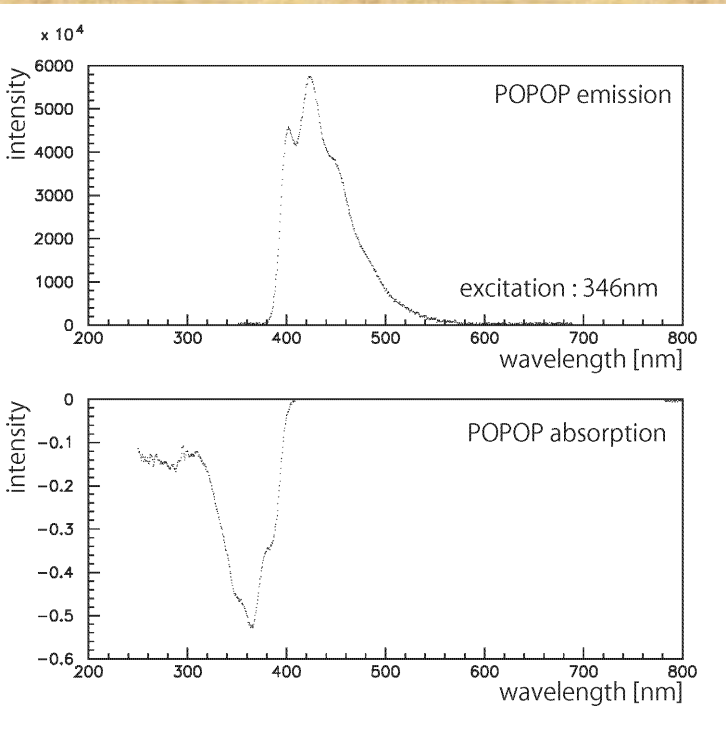
concentration of $Zr(acac)_4$	Observed channel	Expected channel
0 mg	2450	2450
50mg	1800	1997
100mg	1400	1687
200mg	950	1284
300mg	650	1038
500mg	300	750

Photo Luminescence and absorption of PPO



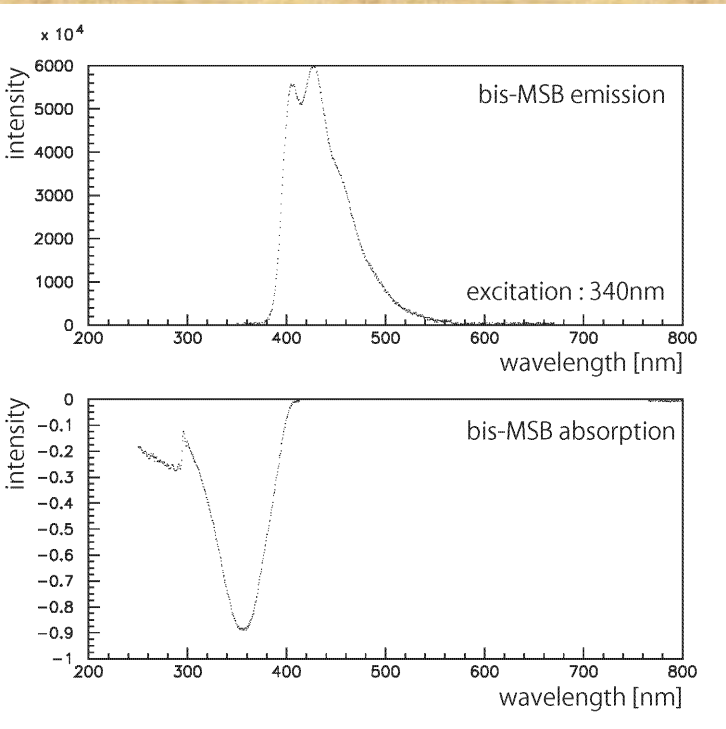
- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance device : HITACHI U-3000
  - Solvent : Benzonitrile (PhCN)
  - Concentration : 1.0×10^{-5} mol/L
  - 2,5-Diphenyloxazole
  - Molecular mass : 221.26
  - Max. emission wavelength : 368.0nm
  - Max. absorption wavelength : 309.7nm

Photo Luminescence and absorption of POPOP



- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance device : HITACHI U-3000
  - Solvent : Benzonitrile (PhCN)
  - Concentration : 1.0×10^{-5} mol/L
  - 1,4-Bis(5-phenyloxazol-2-yl)benzene
  - Molecular mass : 364.40
  - Max. emission wavelength : 423.6nm
  - Max. absorption wavelength : 364.1nm

Photo Luminescence and absorption of bis-MSB



- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance device : HITACHI U-3000
  - Solvent : Benzonitrile (PhCN)
  - Concentration : 1.0×10^{-5} mol/L
  - 1,4-Bis(2-methylstyryl)benzene
  - Molecular mass : 310.44
  - Max. emission wavelength : 426.6nm
  - Max. absorption wavelength : 355.3nm