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実験Ⅳ

日本物理学会　第６９回年次大会

2014年3月27日

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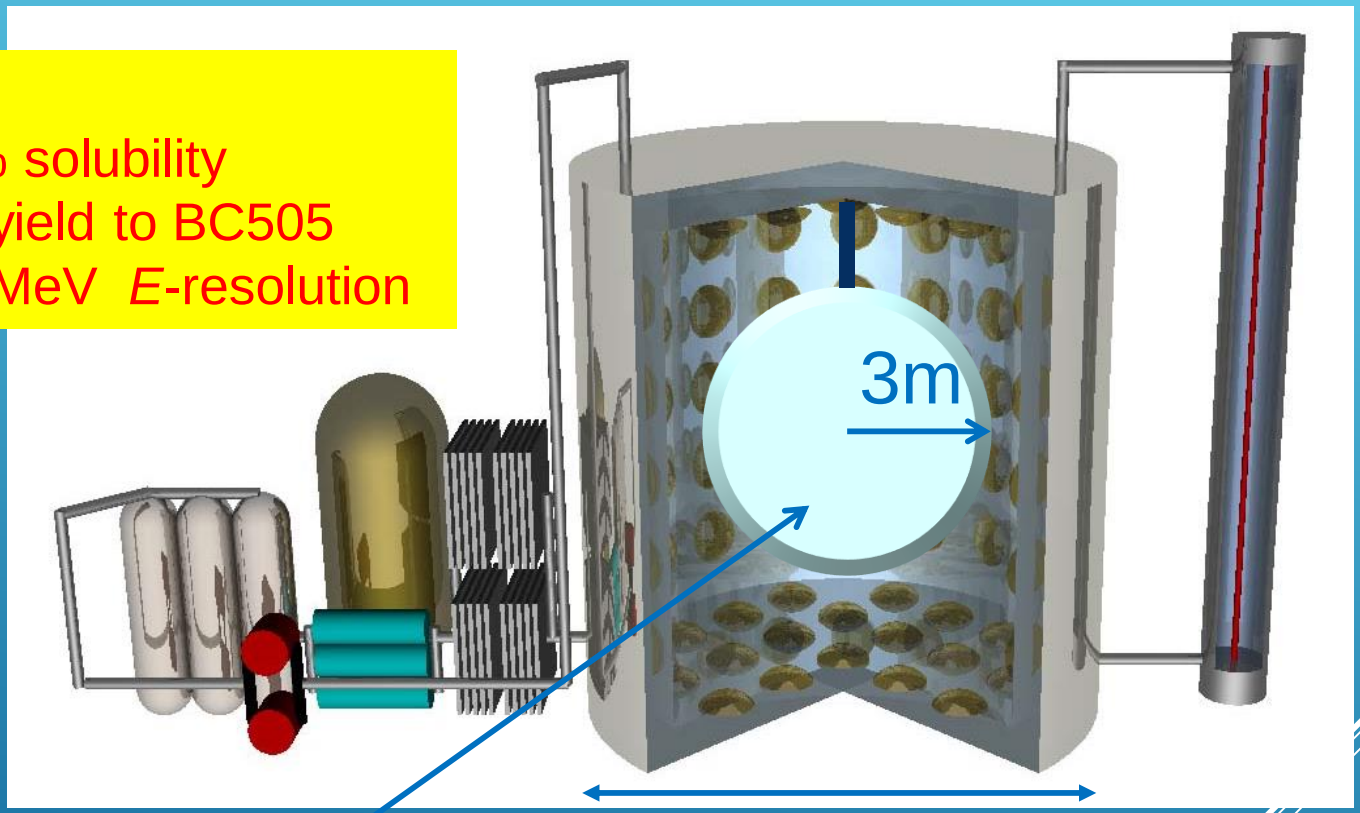
ZIRCONIUM COMPLEX IN ORGANIC LIQUID SCINTILLATOR FOR DOUBLE BETA DECAY EXPERIMENT (ZICOS EXPERIMENT)

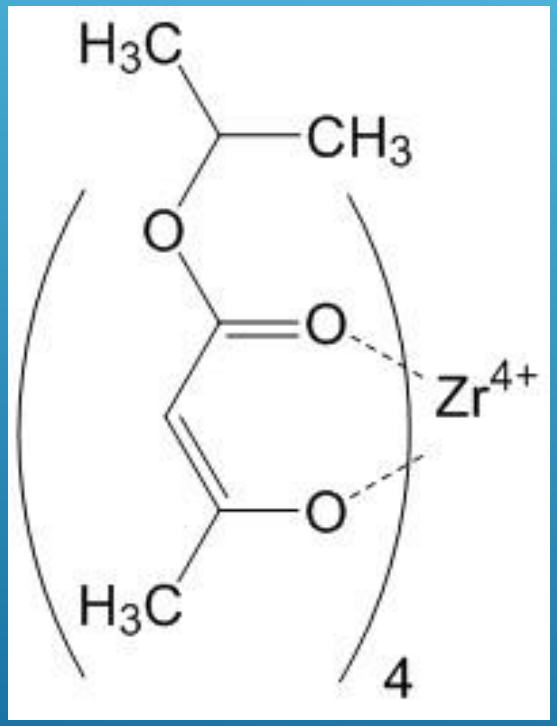
Goals :
(1) ≧ 10w.t.% solubility
(2) 60% light yield to BC505
(3) 4% @ 2.5MeV *E*-resolution



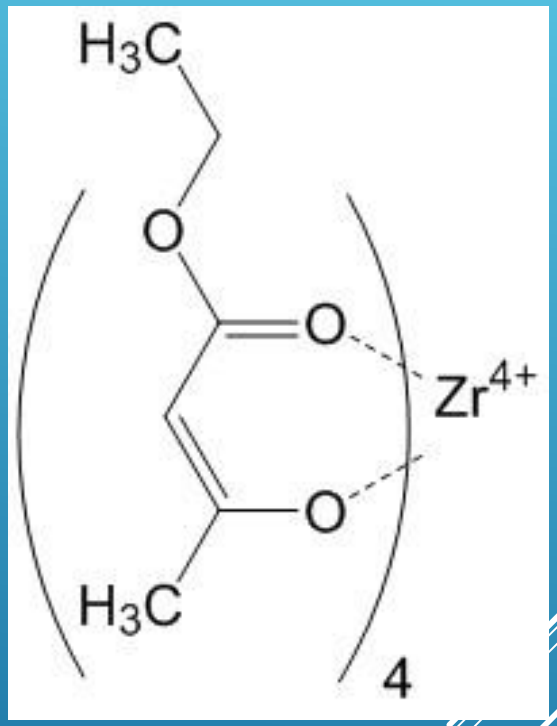
Zirconium loaded 100ton LS
(300kg ^{96}Zr assuming10% enrich)

ZIRCONIUM BETA-KETO ESTER COMPLEX

$Zr(CH_3COCHCOOCH(CH_3)_2)_4$
= $Zr(iprac)_4$
mw : 663.87

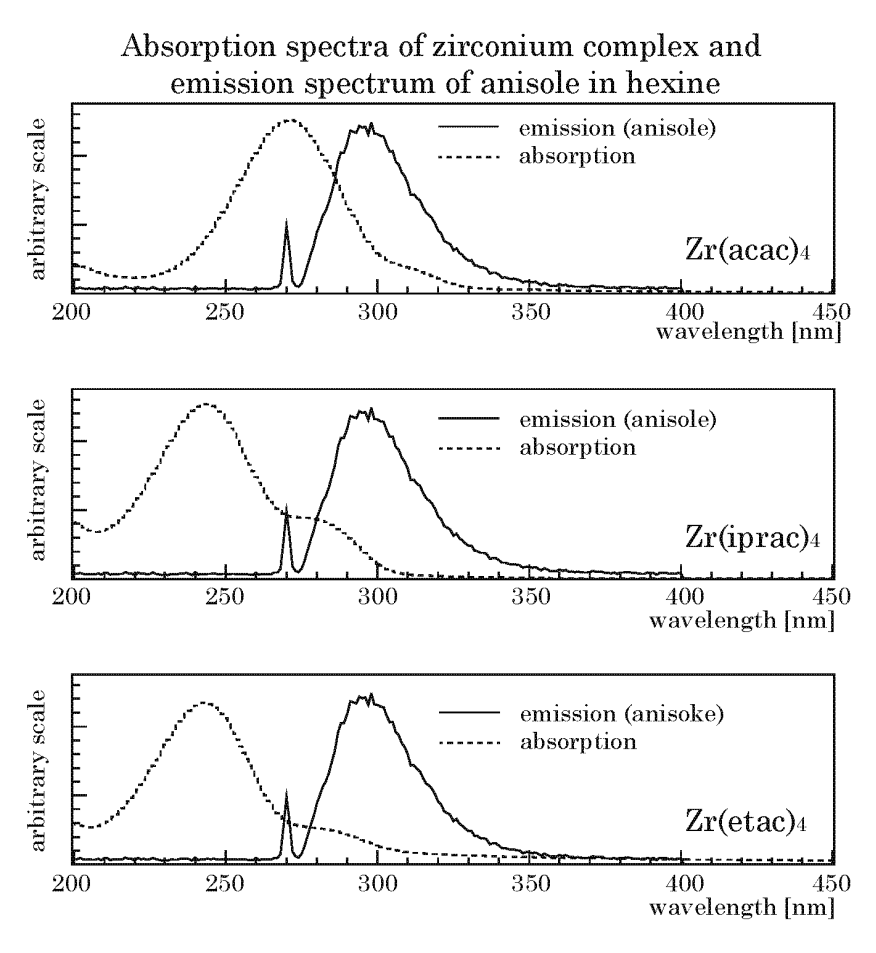


$Zr(CH_3COCHCOOCH_2CH_3)_4$ =
$Zr(etac)_4$
mw : 607.76



Solubility > 10 w.t.% for anisole

ABSORBANCE SPECTRA FOR ZIRCONIUM BETA-KETO ESTER COMPLEX



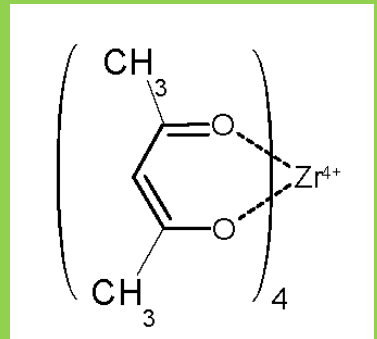
Absorption peaks of zirconium β-keto ester complex were found around at 245nm, however small bump existed around 280nm.

$Zr(CH_3COCHCOCH_3)_4$ =

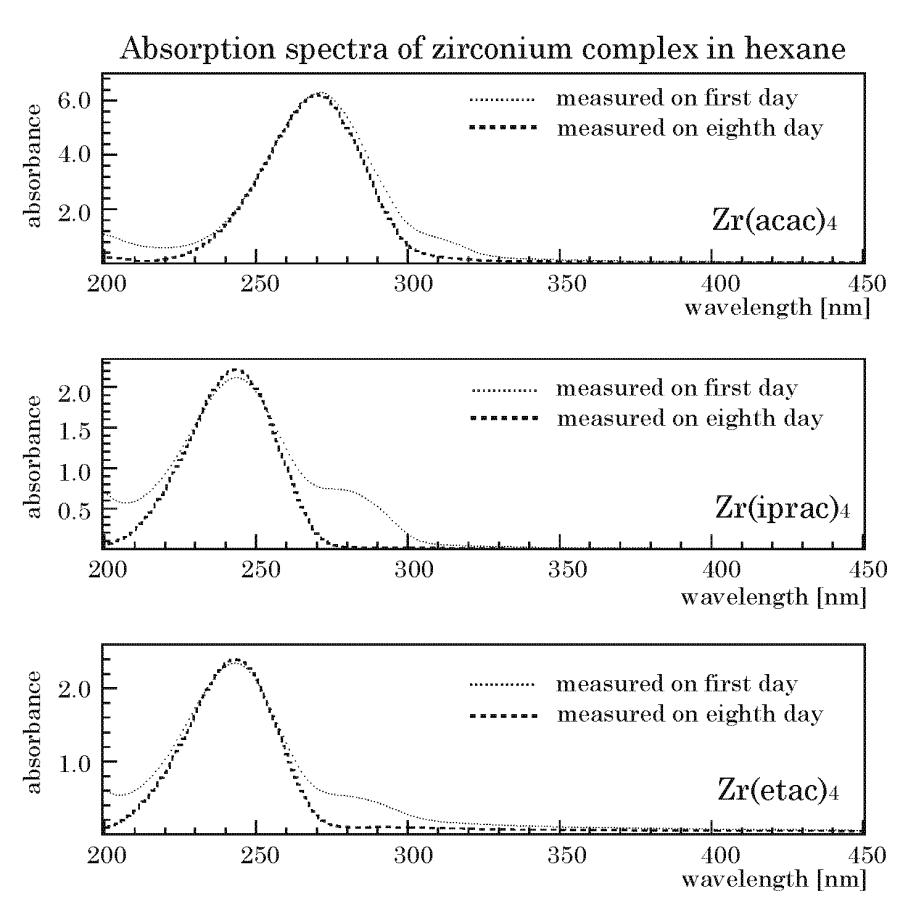
$Zr(acac)_4$

mw :

487.66



ABSORBANCE SPECTRA FOR ZIRCONIUM BETA-KETO ESTER COMPLEX AFTER 1 WEEK

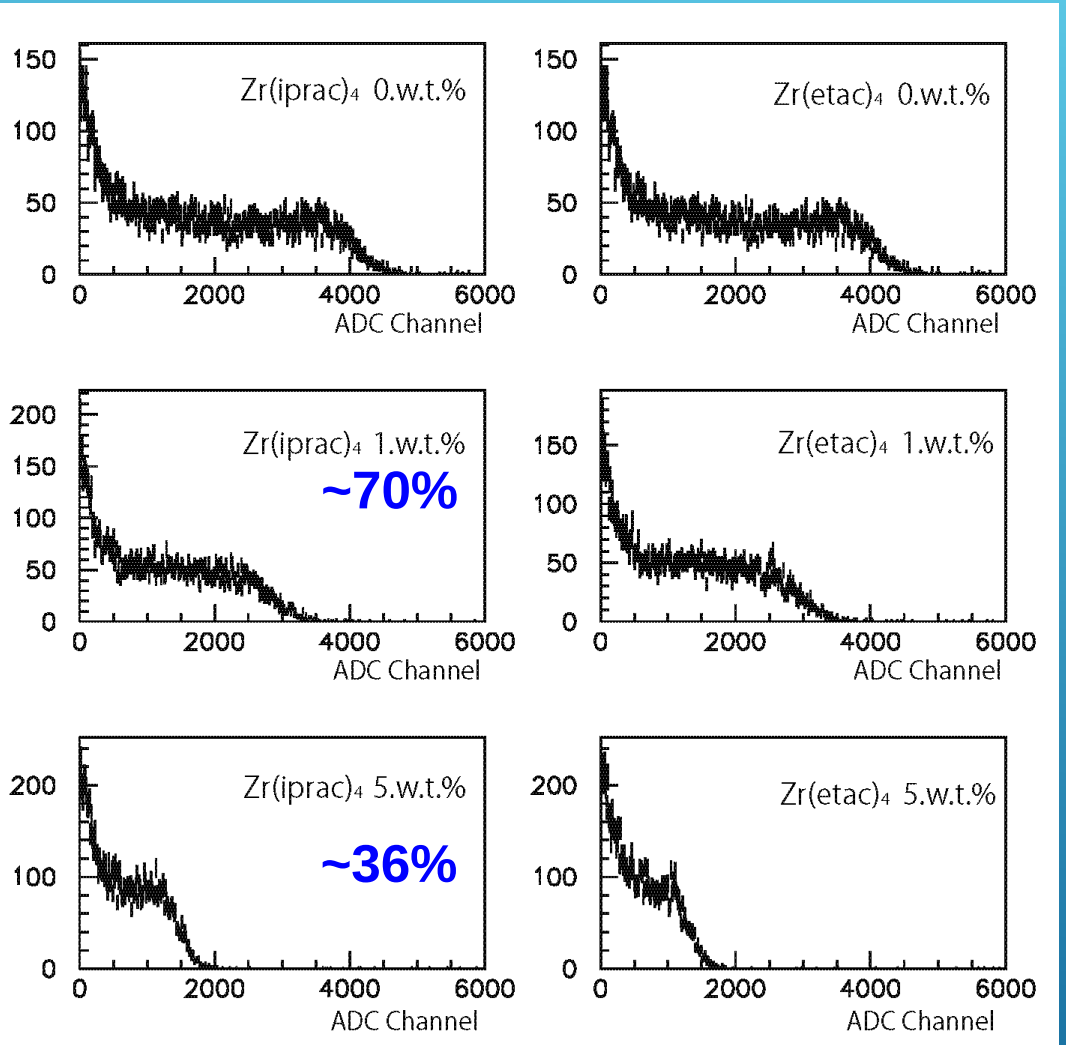


Small bump disappeared after ~ 1 week.

It could be explained by impurities of Zr complex, and they should be precipitated on bottom of the vial.

(see explanation slide)

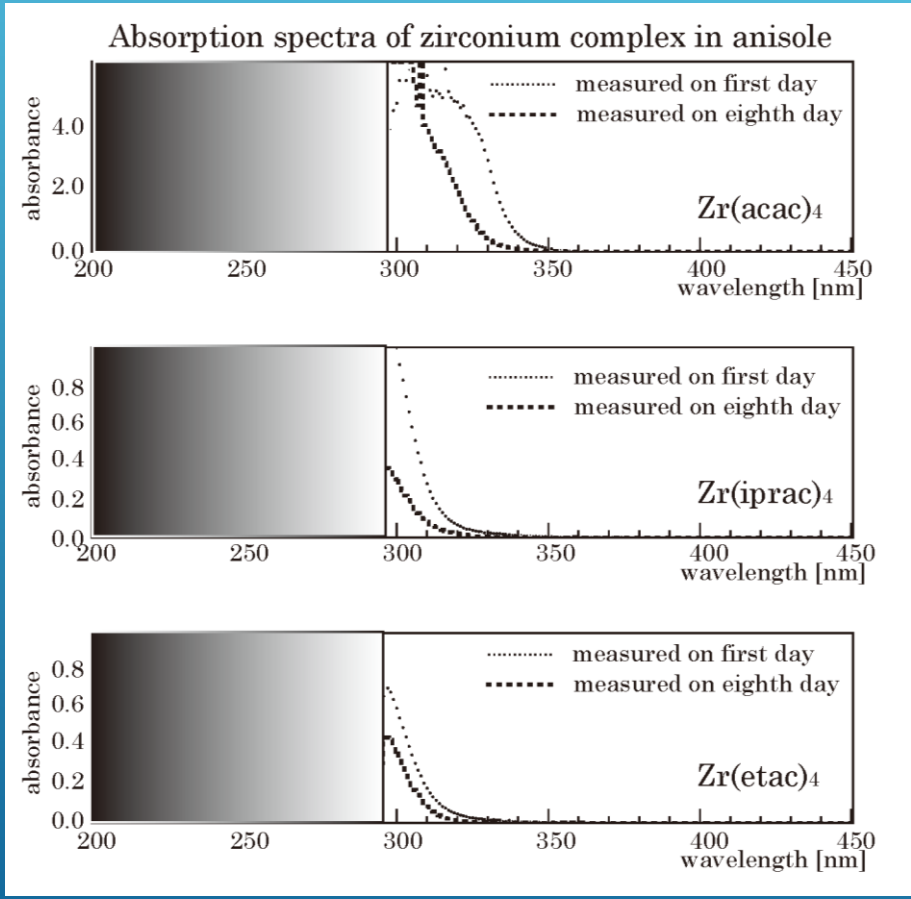
LIGHT YIELD OF LS CONTAINING ZIRCONIUM BETA-KETO ESTER COMPLEX



Light yield decreased as increasing the concentration of the complex.

Still exists small bump of absorption around 280nm in anisole ?

ABSORBANCE SPECTRA FOR ZIRCONIUM BETA-KETO ESTER COMPLEX IN ANISOLE

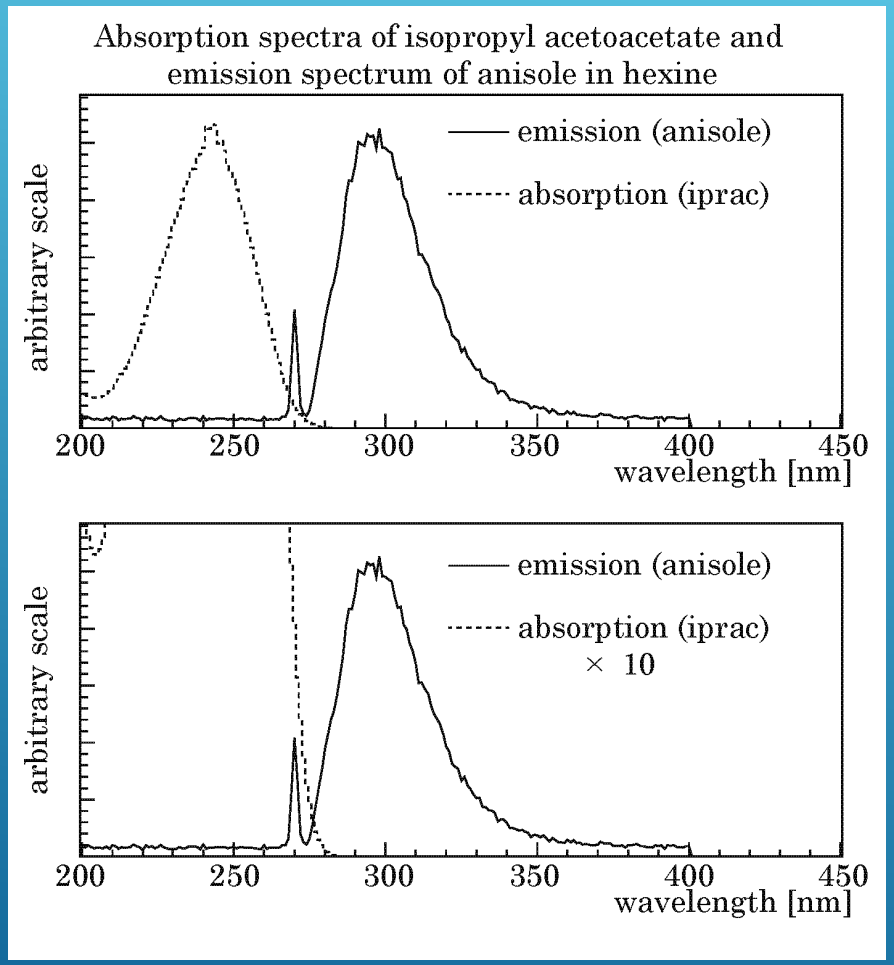


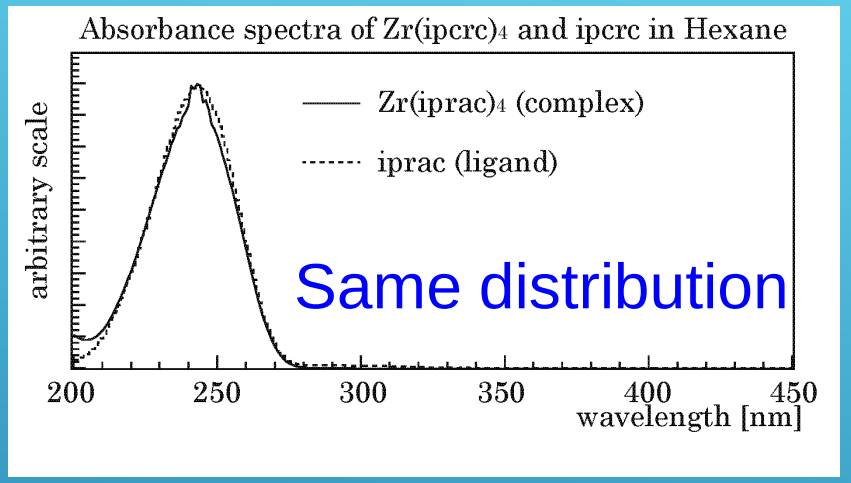
Small bump around 280nm in anisole did not disappear even after ~ 1 week.

Impurities might be dissolved in anisole…

We have to purify the complex by using such as sublimation.

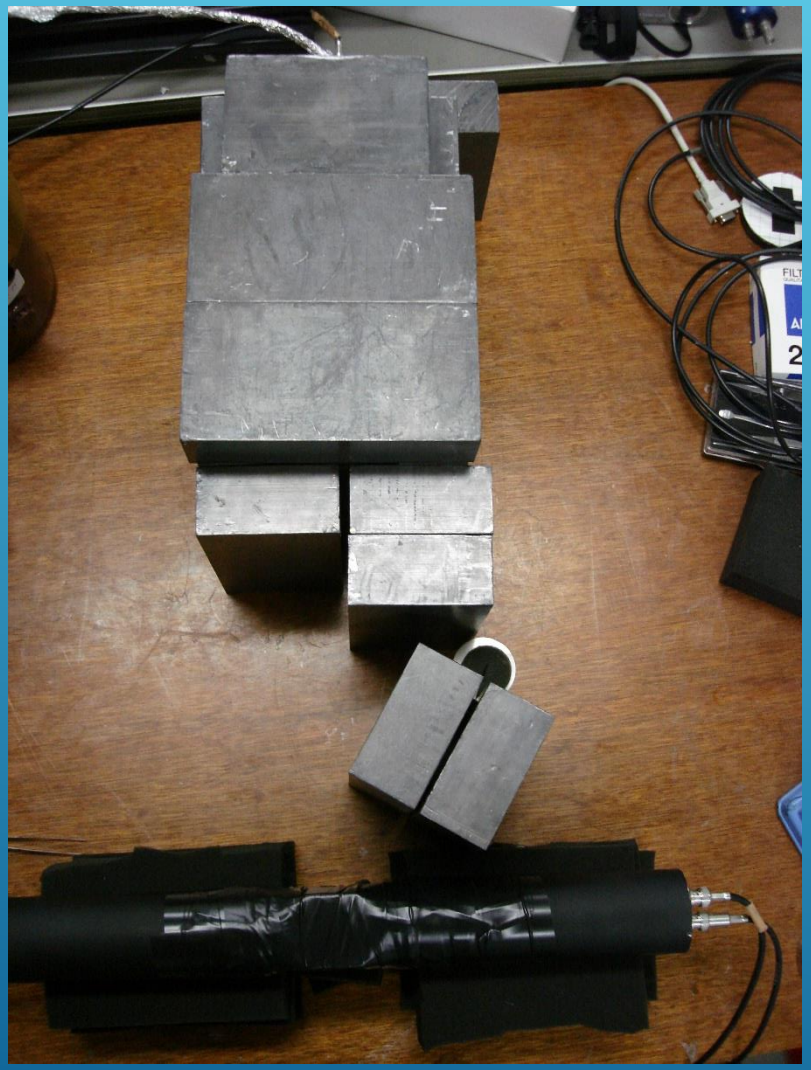
ABSORBANCE SPECTRA FOR ZIRCONIUM BETA-KETO ESTER COMPLEX AND LIGANDS

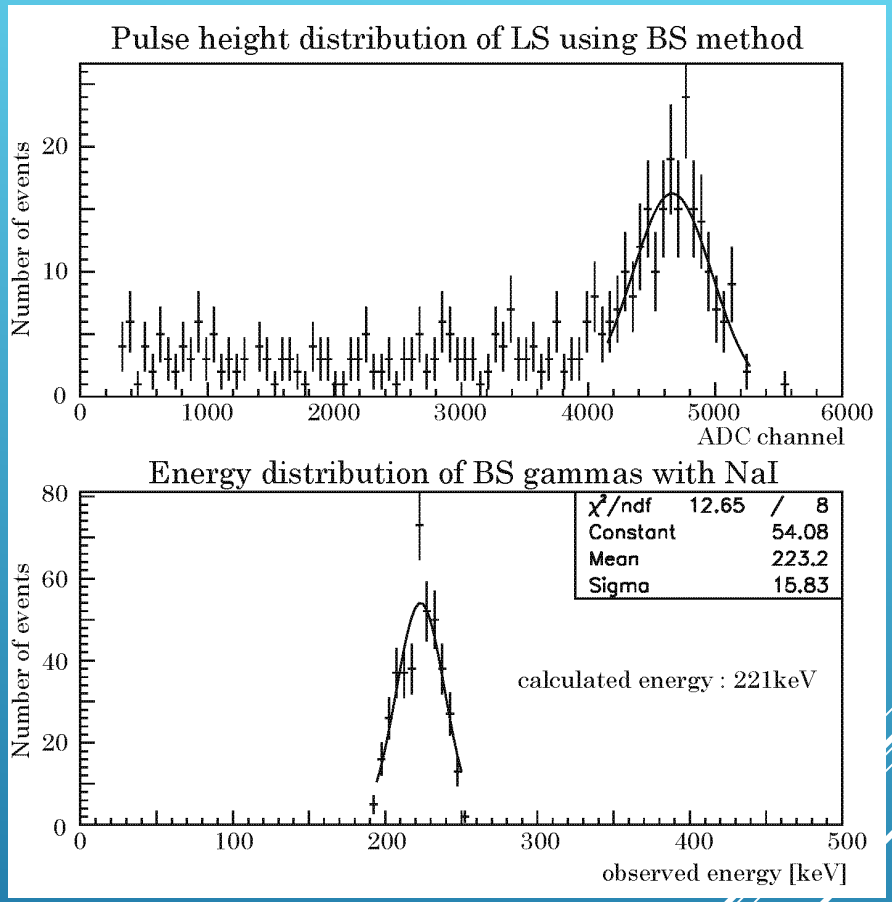




Absorption tail of β-keto ester ligands slightly overlapped with the region of the emission of anisole.

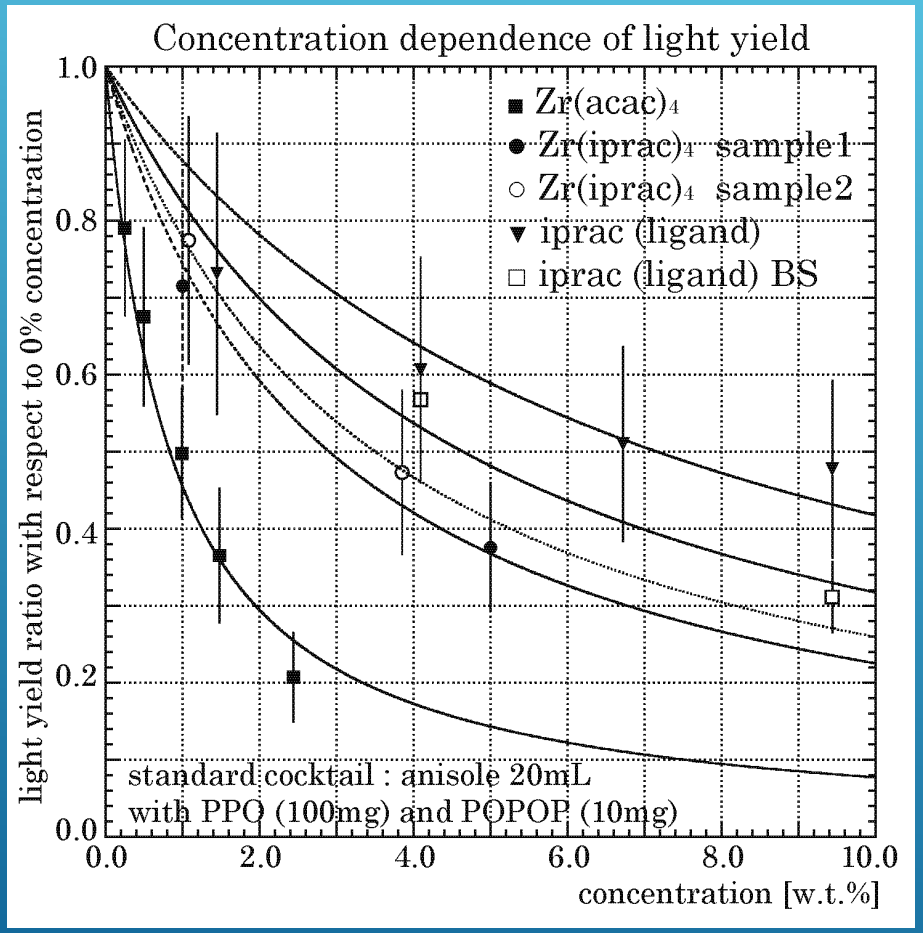
BACK SCATTERING METHOD





Single peak could be used even in liquid scintillator.

LIGHT YIELD OF LS CONTAINING ZIRCONIUM COMPLEX AS A FUNCTION OF CONCENTRATION



Light yield of $Zr(iprac)_4$ even with small bump recovered about double compared with $Zr(acac)_4$.

Light yield at 10w.t.% concentration was almost 40% to BC505 (≒ standard cocktail) , If the small bump could be removed.

ENERGY RESOLUTION OF LS CONTAINING ZIRCONIUM COMPLEX AS A FUNCTION OF CONCENTRATION

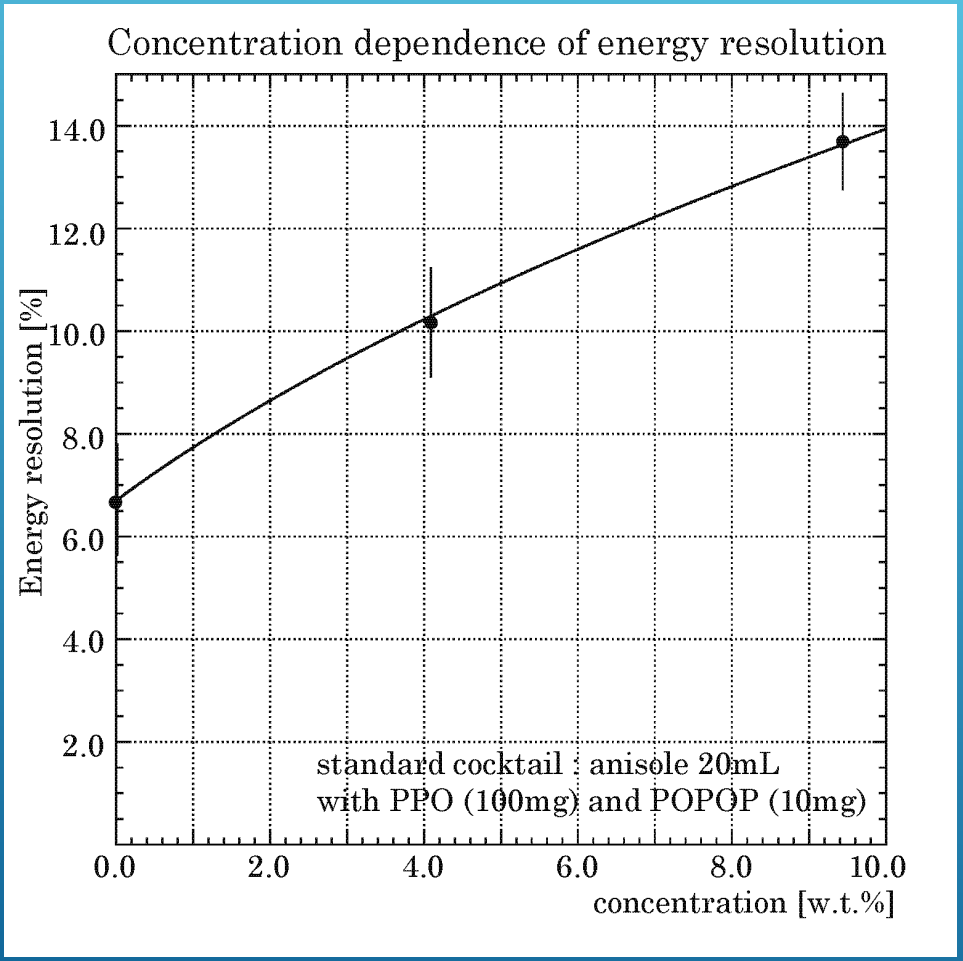


Photo coverage : ~8.5% (see explanation slide) Assuming 40% of photo coverage, the energy resolution will recover 6.5% @ 1MeV = 4.1% @ 2.5MeV for 10 w.t.% concentration.

They almost achieved to our goal!.

SUMMARY

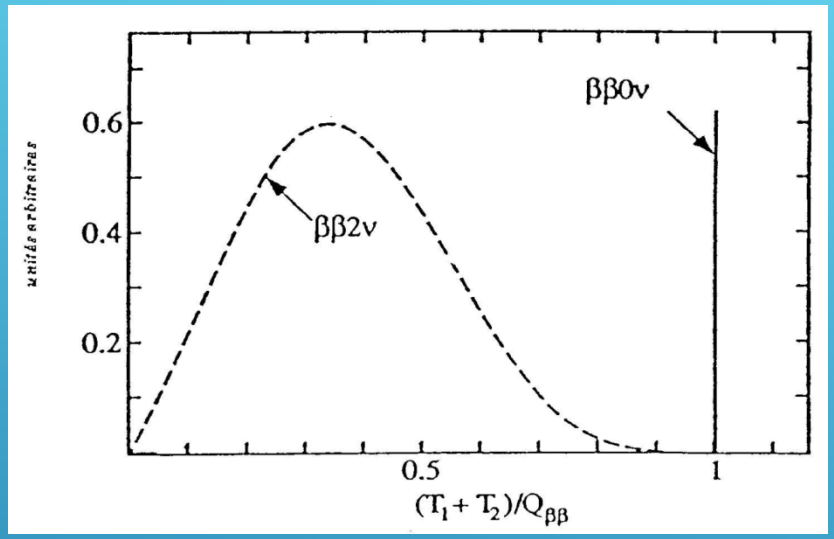
- Confirmed that the absorption peak moved shorter wavelength (275nm → 245nm) by introducing β-keto ester substituent groups.
- Small bump was found in absorption spectra. They could be explained by impurities of complex, and would be removed by purifying complex such as sublimation.
- Anisole based liquid scintillator with 10 w.t.% concentration of $Zr(iprac)_4$ possibly has both 40% for light yield to BC505 and 4.1%@2.5MeV (assuming 40% photo coverage) for energy resolution, so that they almost achieved our goal!
- To improve light yield (and also energy resolution), we shall move the absorption peak around 210nm using $Zr(deml)_4$ "tetrakis (diethyl malonato) zirconium".

BACKUP

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: target mass

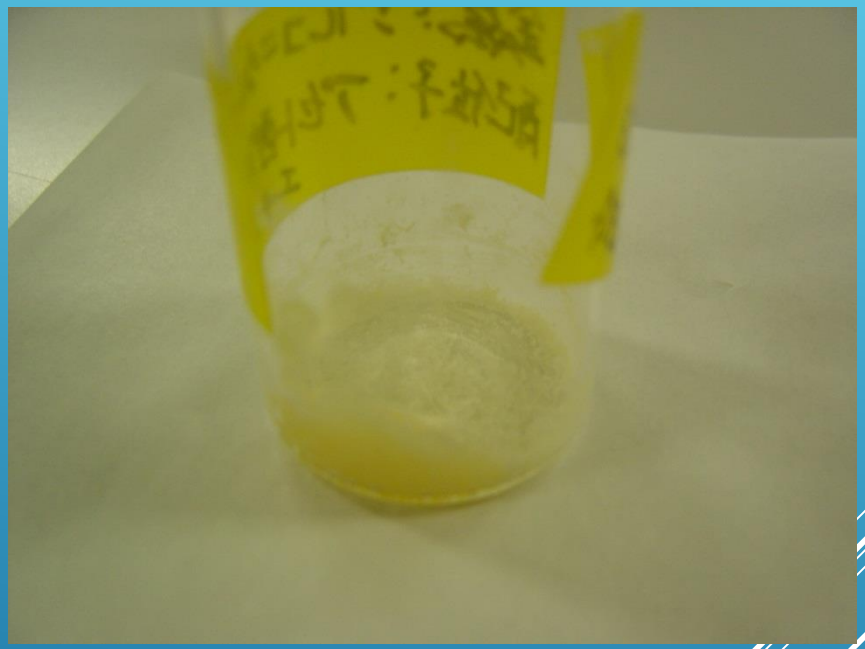
t: measurement time ΔE: energy resolution B: BG rate

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

SYNTHESIZE OF ZIRCONIUM BETA-KETO ESTER COMPLEX

$Zr(iprac)_4$
state: powder

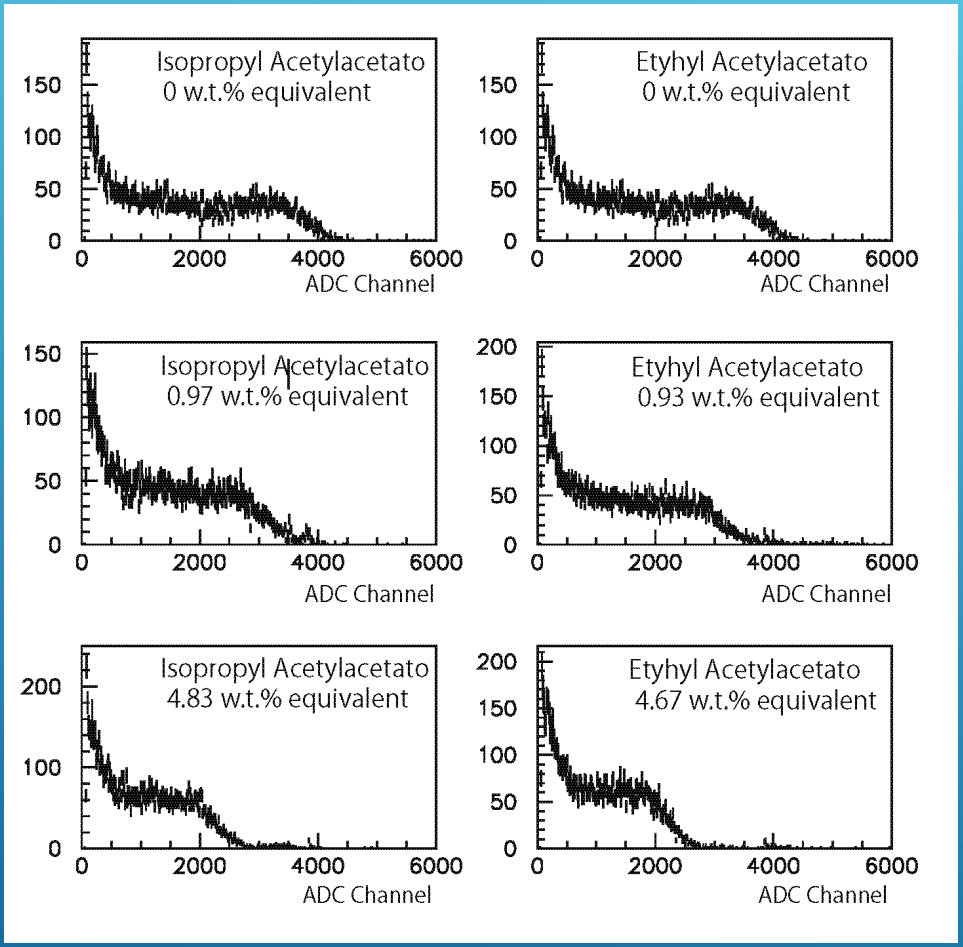
$Zr(etac)_4$
state : dry solid



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

Solubility > 10 w.t.% for anisole

LIGHT YIELD OF LS CONTAINING BETA-KETO ESTER LIGAND

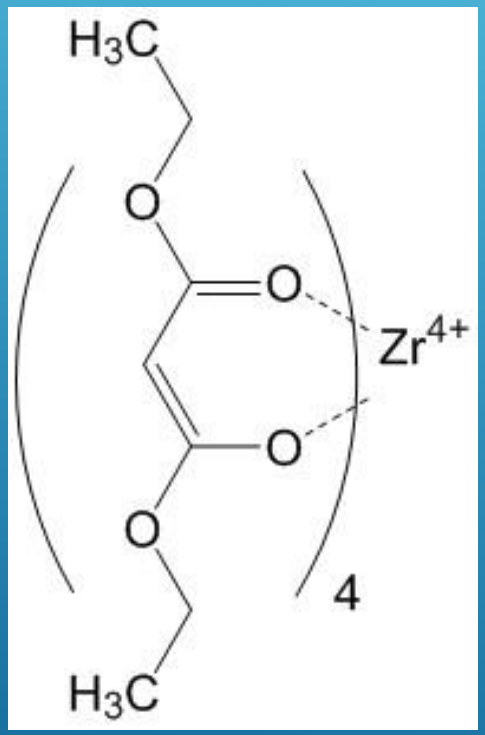


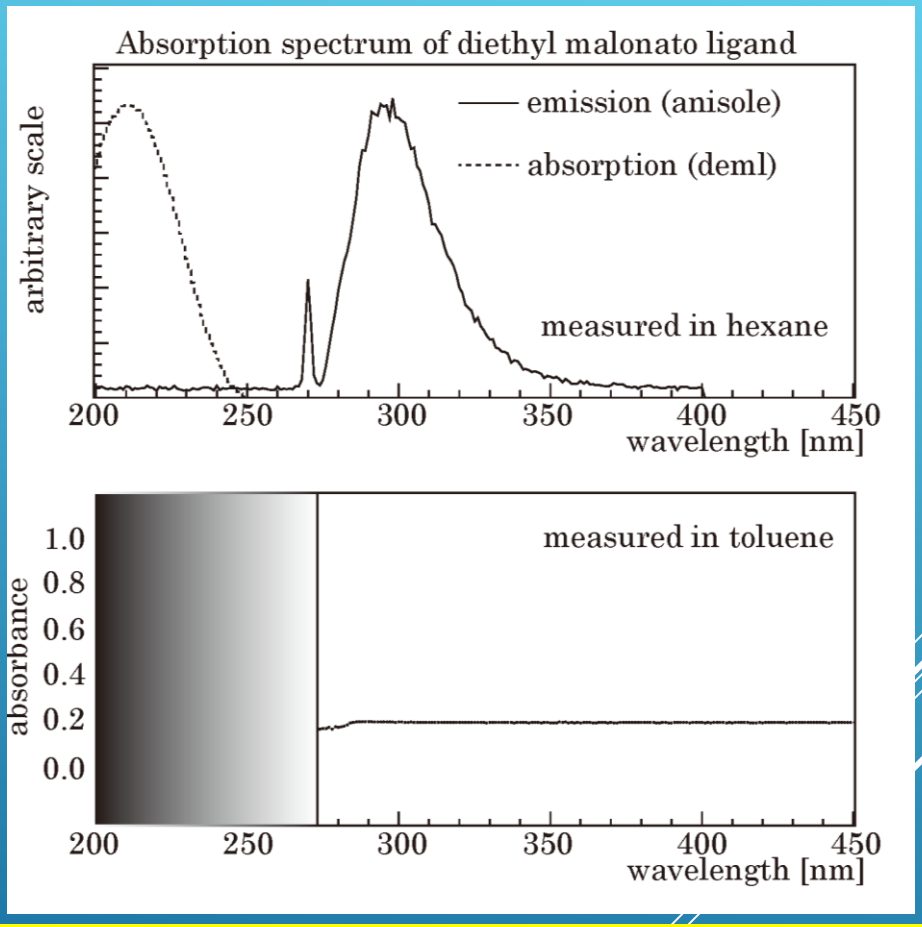
Light yield recovered about 50% compared to the Zr-keto ester complex due to vanish the small bump.

However, a quenching occurred due to the absorption tail.

TETRAKIS (DIETHYL MALONATO) ZIRCONIUM AND ABSORBANCE SPECTRUM OF LIGAND

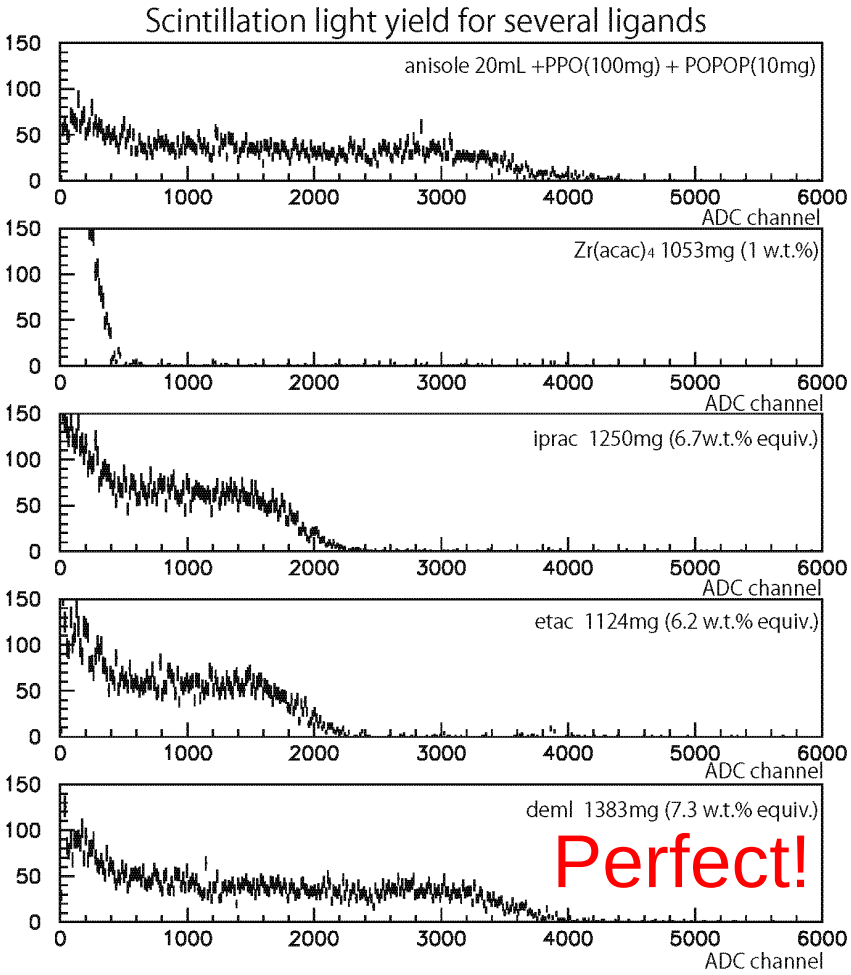
$Zr(CH_3CH_2OCOCHCOOCH_2CH_3)_4$
$= Zr(deml)_4$ mw : 727.87





shorter wavelength (~210nm)

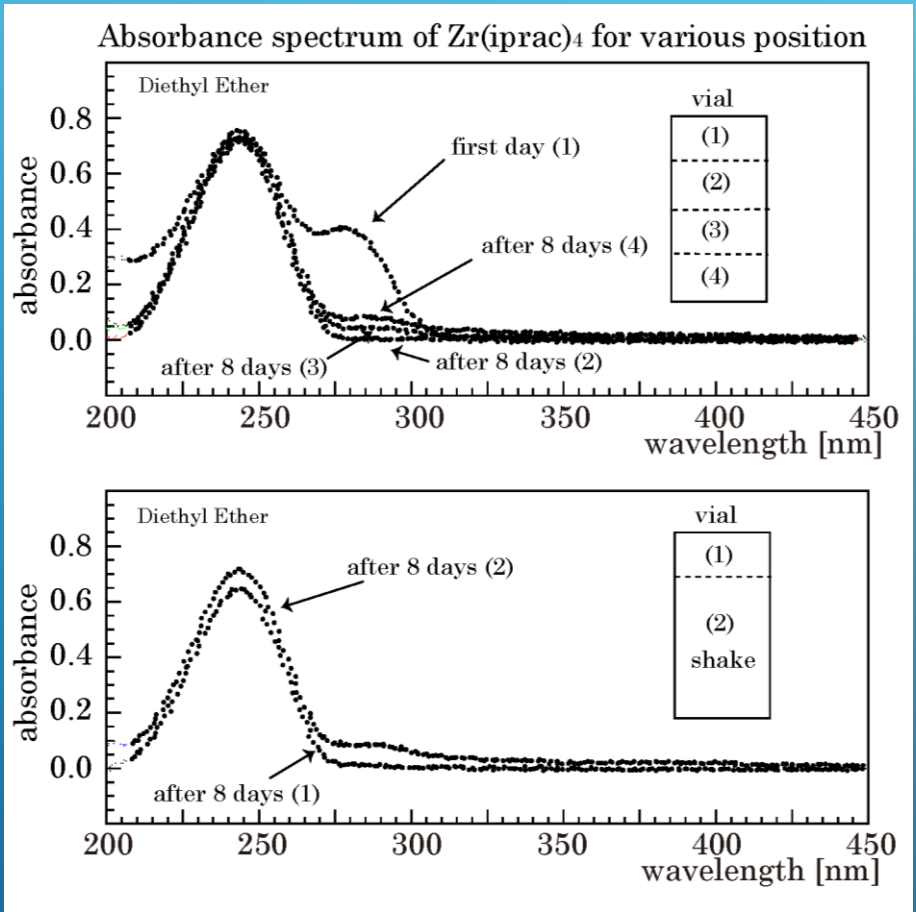
LIGHT YIELD OF LS CONTAINING DIETHYL MALONATO LIGAND



No quenching due to overlap between the absorption of ligand and the emission of anisole should be occurred.

$Zr(deml)_4$ will be an ultimate complex if the solubility becomes over 10w.t.% for anisole.

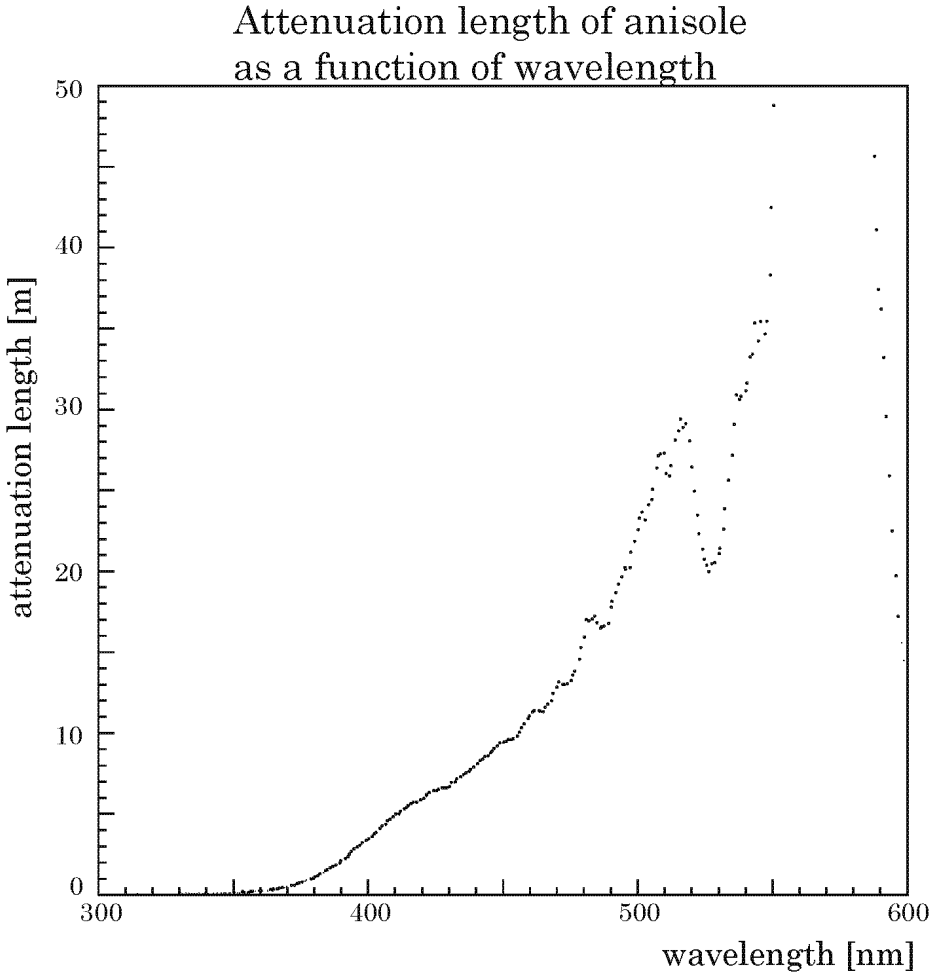
ABSORBANCE SPECTRA OF ZIRCONIUM BETA-KETO ESTER COMPLEX FOR VARIOS POSITION



Small bump disappeared after ~ 1 week.

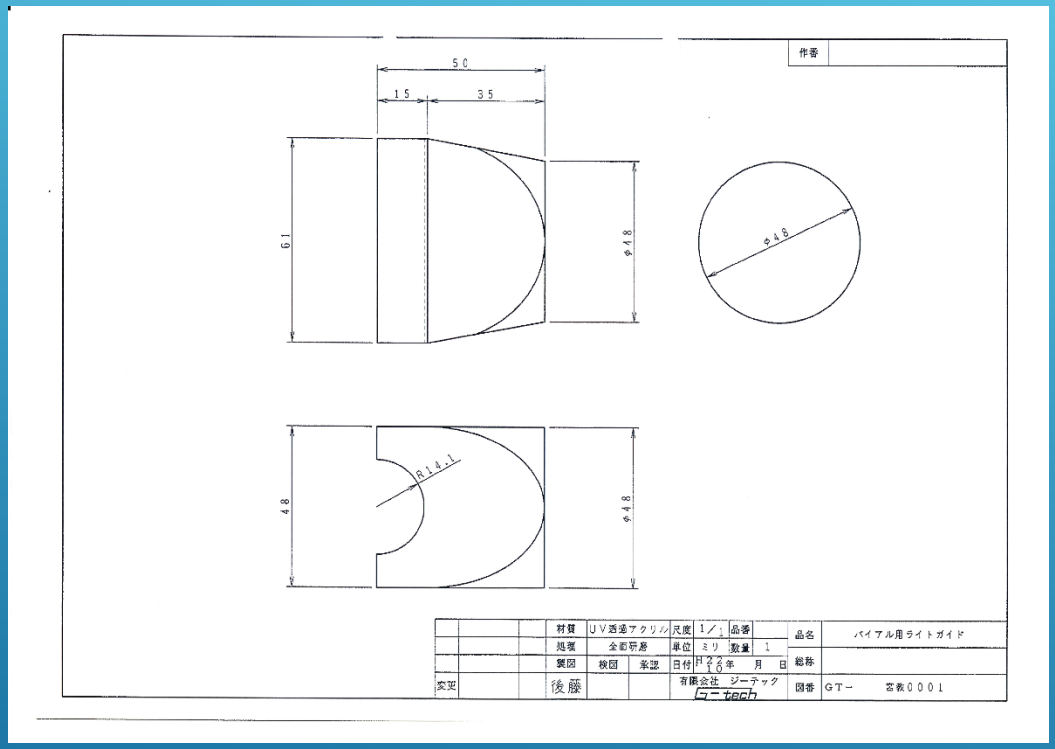
These could be some impurities of Zr complex and they precipitated on bottom of the vial.

ATTENUATION LENGTH OF ANISOLE



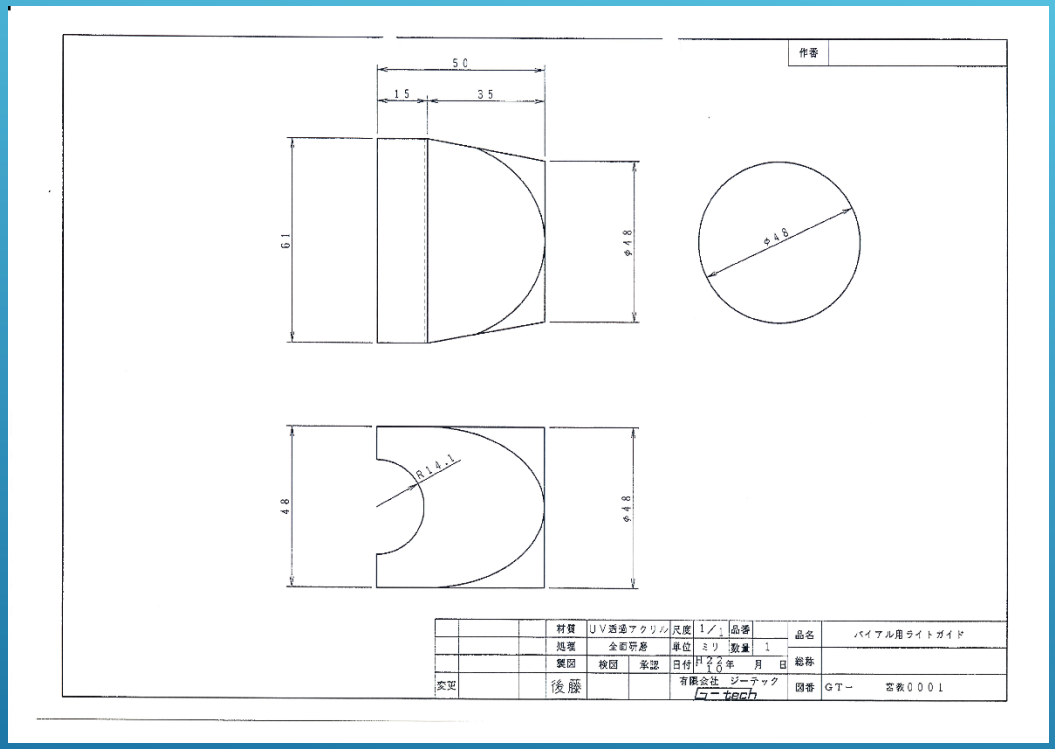
Attenuation length of light from POPOP was obtained as ~6m. It is almost equivalent with the detector size.

PHOTO COVERAGE



Scintillation light was guided to PMT via the acrylic light guide. From the computer simulation, the solid angle was obtained as 8.5% for 4π area.

PHOTO COVERAGE



Scintillation light was guided to PMT via the acrylic light guide. From the computer simulation, the solid angle was obtained as 8.5% for 4π area.

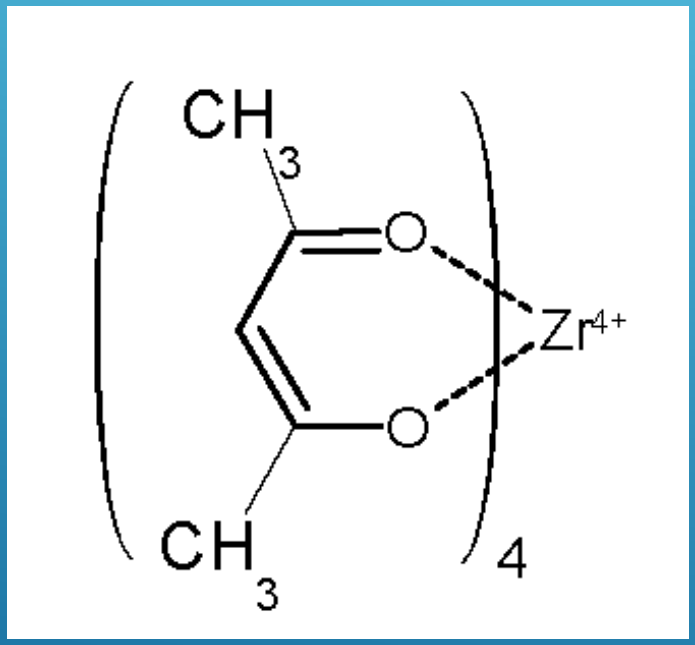
Light yield calculation

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

L_0 : Light yield of anisole
N_{ppo} : No. of PPO molecular
N_{Zr} : No. of $Zr(acac)_4$ molecular
σ_1: absorbance of PPO
σ_2 : absorbance of complex

ZIRCONIUM B-DIKETON COMPLEX

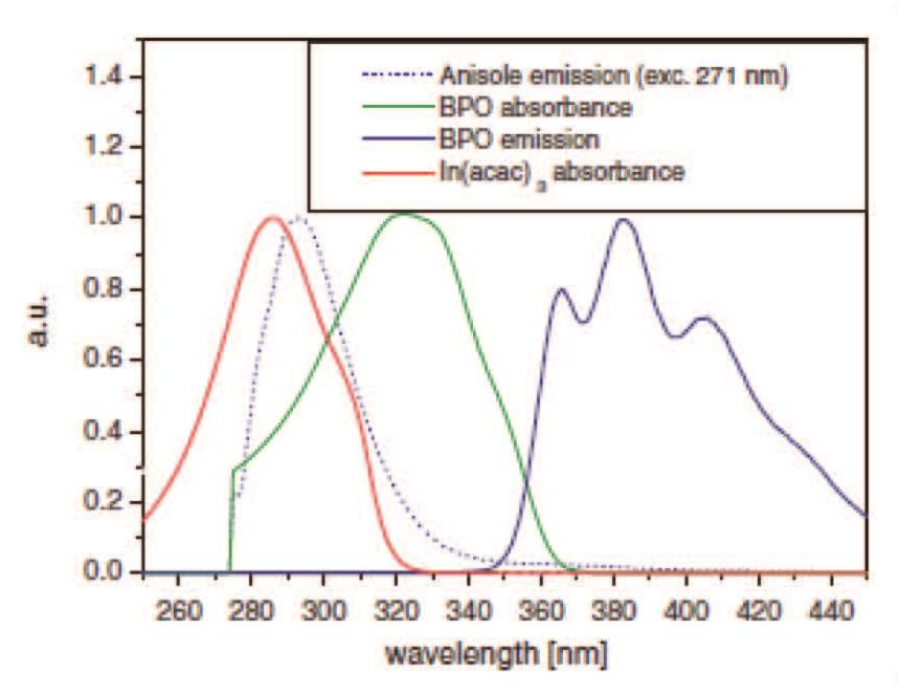
- Zirconium(IV) acetylacetonate ($Zr(acac)_4$)



Molecular weight : 487.66

- Advantage
  - good solubility (over 10w.t.%) in Anisole (PhOMe)
  - Stable and cheep
  - Commercial product
- Disadvantage
  - Low scintillation light yield due to overlap the absorption of ligand and emission of anisole.

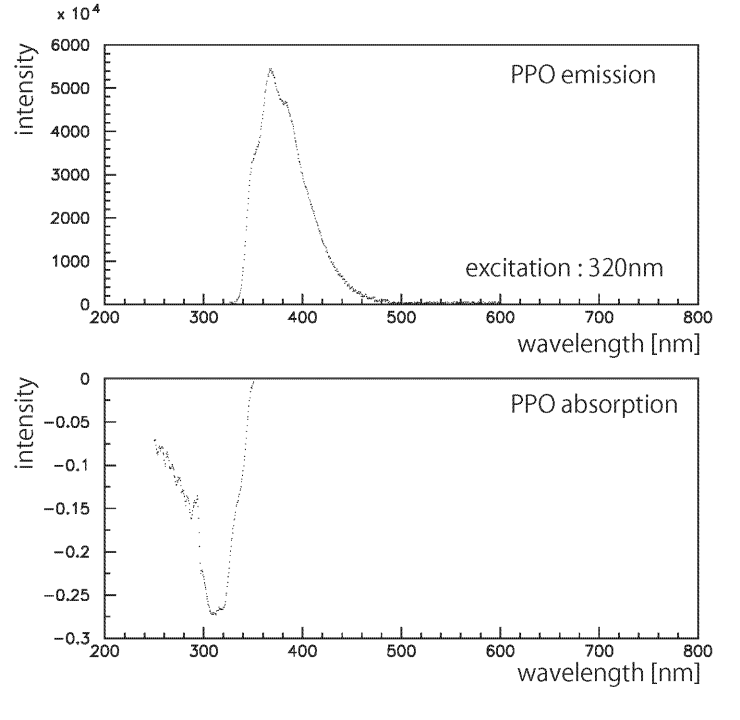
WHAT'S PROBLEM



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

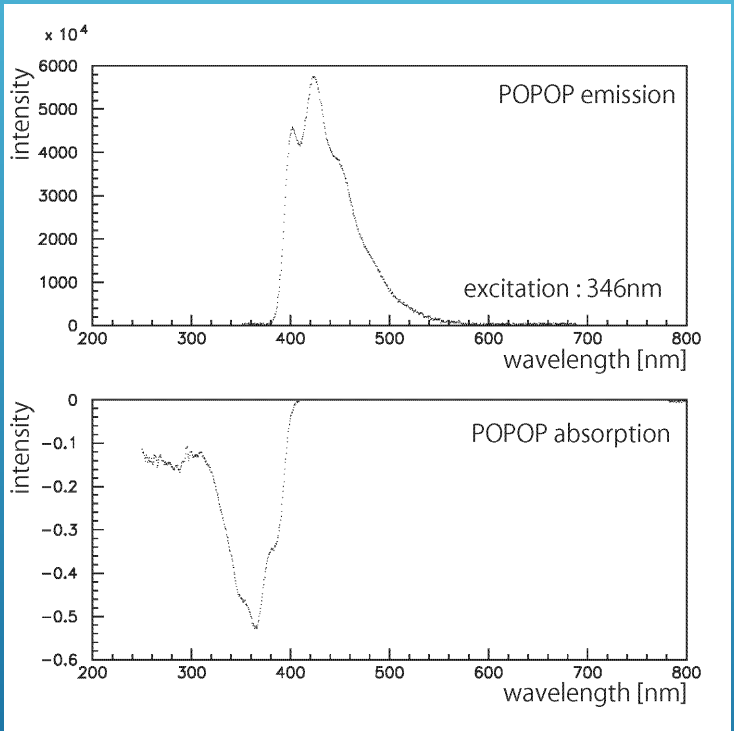
Same overlap between the emission and the absorption could be occurred even if different metal (Zr) was used.

PHOTO LUMINESCENCE AND ABSORPTION OF PPO



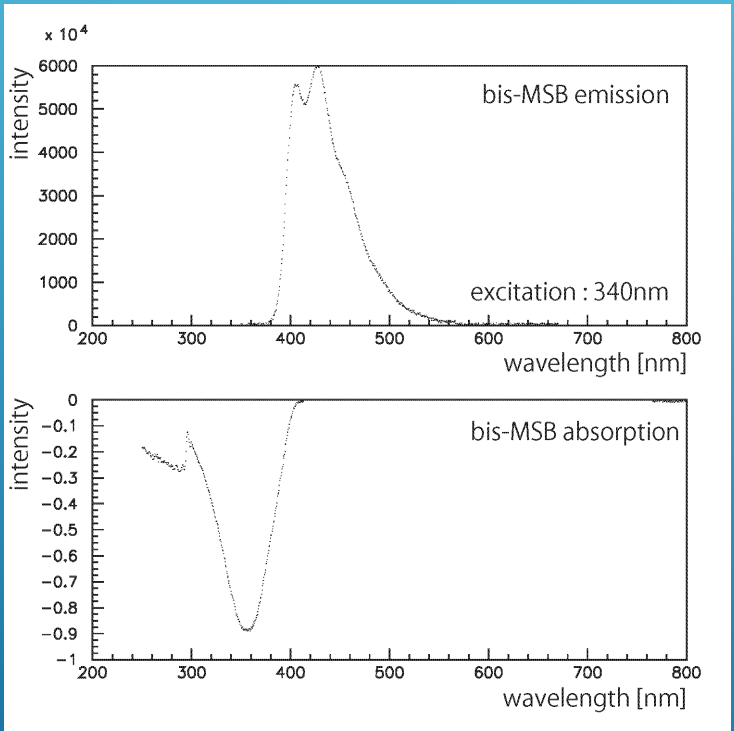
- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance device : HITACHI U-3000
  - Solvent : Benzonitrile (PhCN)
  - Concentration : 1.0×10^{-5} mol/L
  - 2,5-Diphenyloxazole
  - Molecular mass : 221.26
  - Max. emission wavelength : 368.0nm
  - Max. absorption wavelength : 309.7nm

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

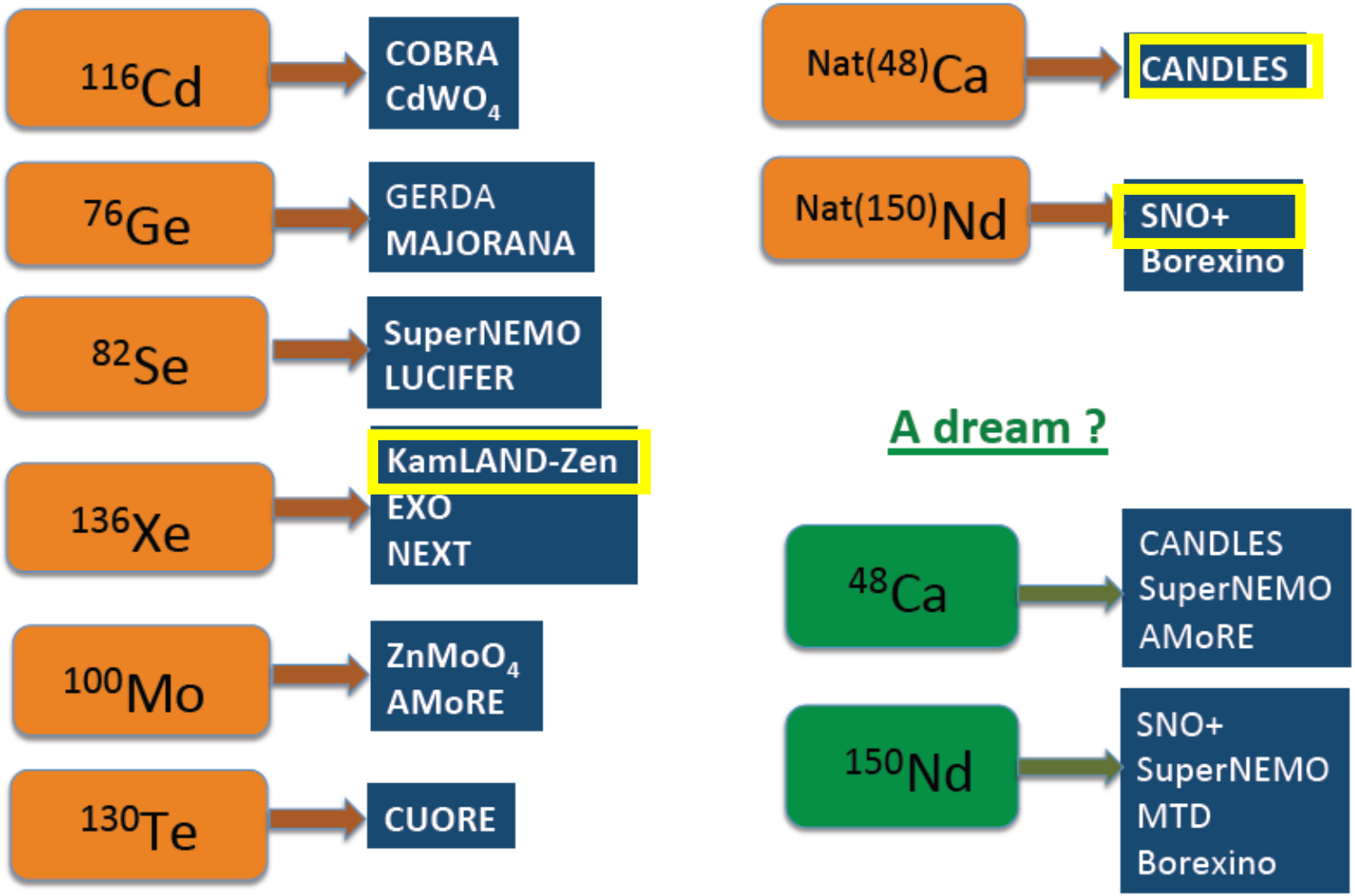
PHOTO LUMINESCENCE AND ABSORPTION OF BIS-MSB



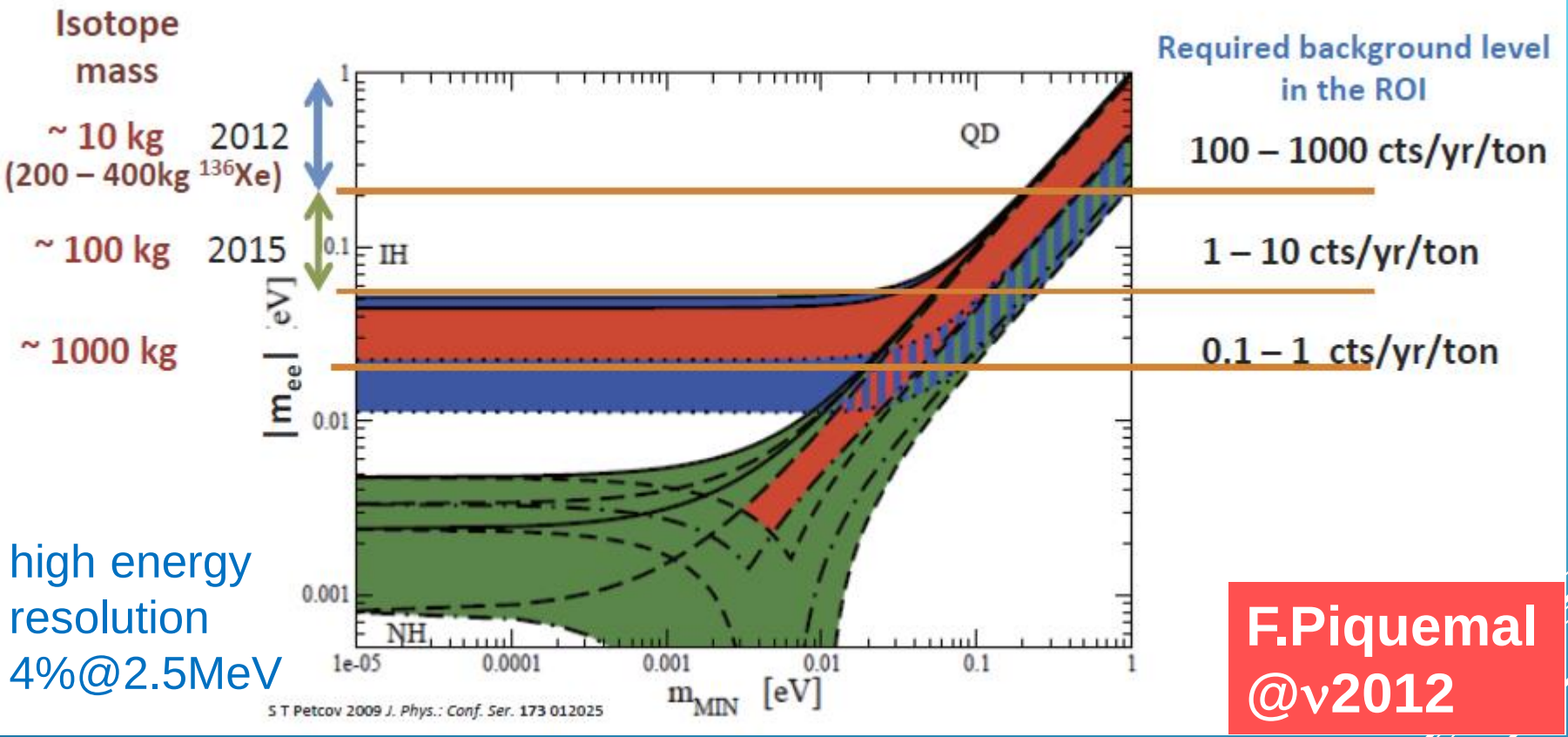
- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance device : HITACHI U-3000
  - Solvent : Benzonitrile (PhCN)
  - Concentration : 1.0×10^{-5} mol/L
  - 1,4-Bis(2-methylstyryl)benzene
  - Molecular mass : 310.44
  - Max. emission wavelength : 426.6nm
  - Max. absorption wavelength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

Studied isotopes

Piquemal@ν2012



FOR FUTURE EXPERIMENTS



high energy resolution 4%@2.5MeV

F.Piquemal @ν2012

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

~tons of target will be necessary for next generation detector