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ジルコニウム９６を用いたニュートリノを放出しない二重ベータ崩壊の研究

新学術領域　「宇宙の歴史をひもとく地下素粒子原子核研究」
第一回　研究会

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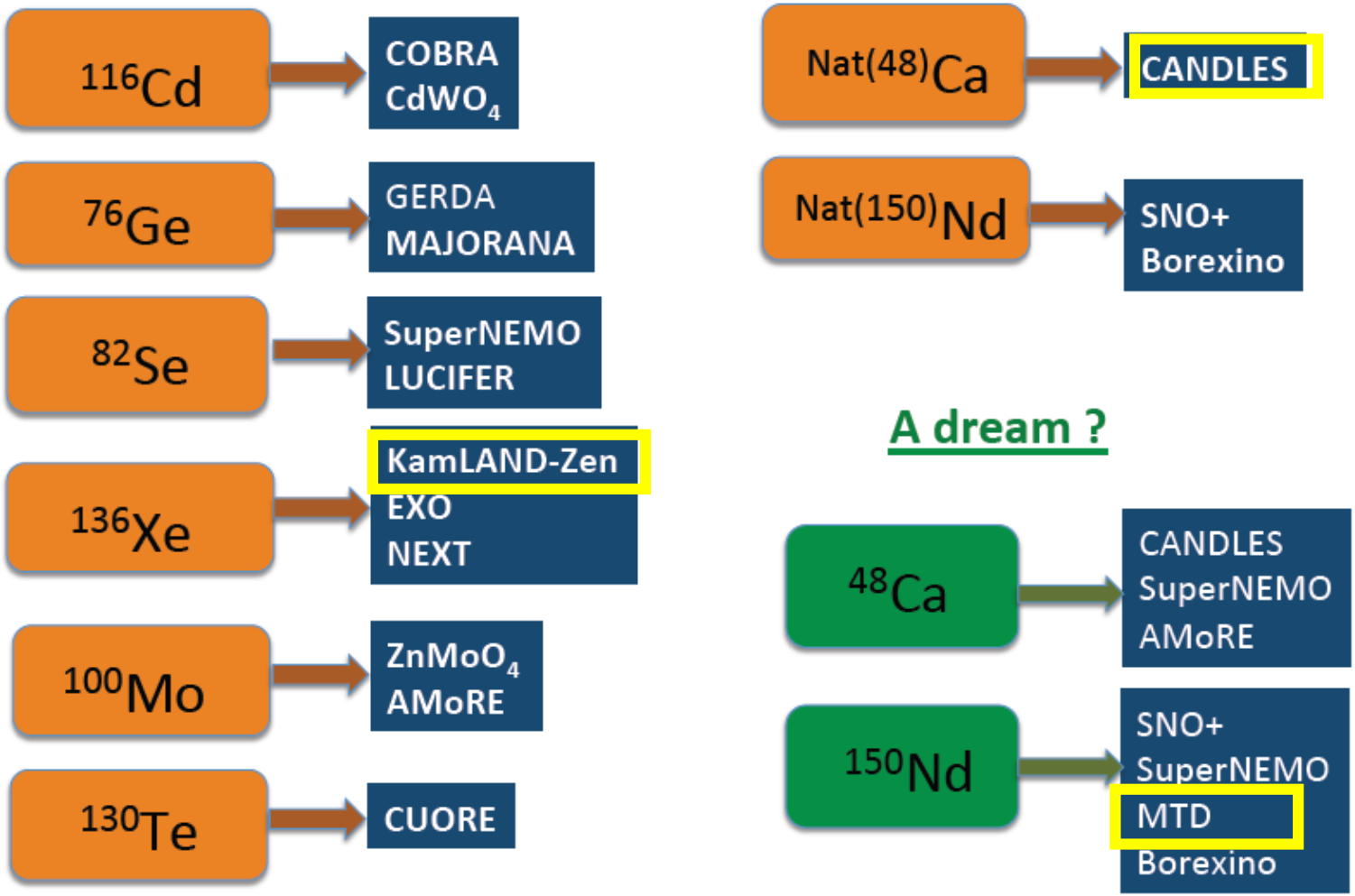
東京理科大学理工学部　郡司天博、塚田　学

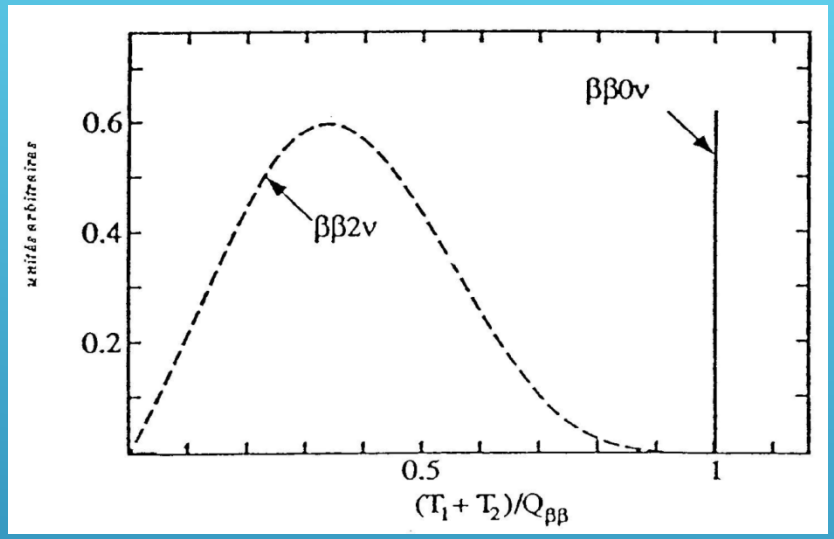
Studied isotopes



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

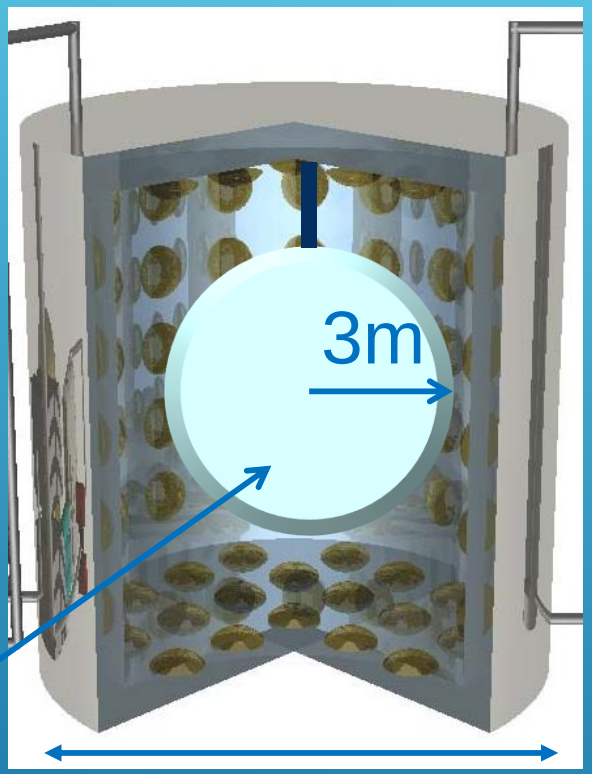
t: measurement time ΔE: energy resolution B: BG rate

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

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

Goals :
(1) Over 10w.t.% solubility
(2) 60% light yield to BC505
(3) 4% at 2.5MeV energy-resolution

Water shield surrounding inner balloon to veto γ's

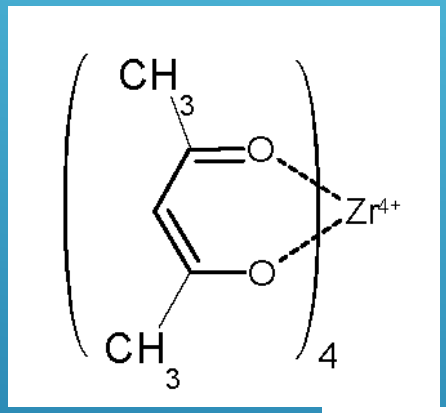


PMT with 40% photo coverage

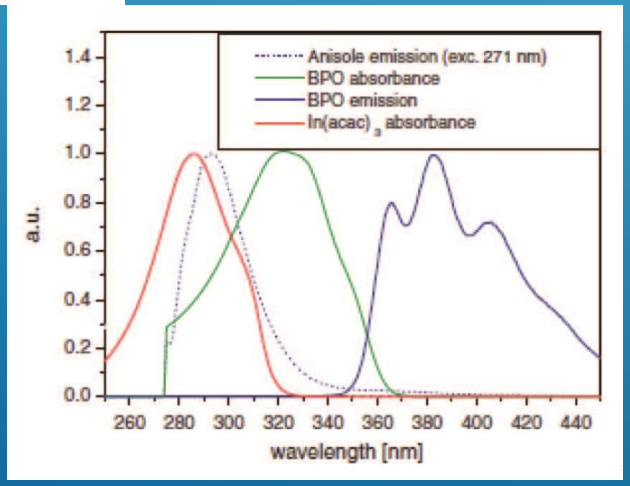
Zirconium loaded 100 tons LS
(300kg ^{96}Zr assuming10% enrich)

ZIRCONIUM BETA-DIKETON COMPLEX

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

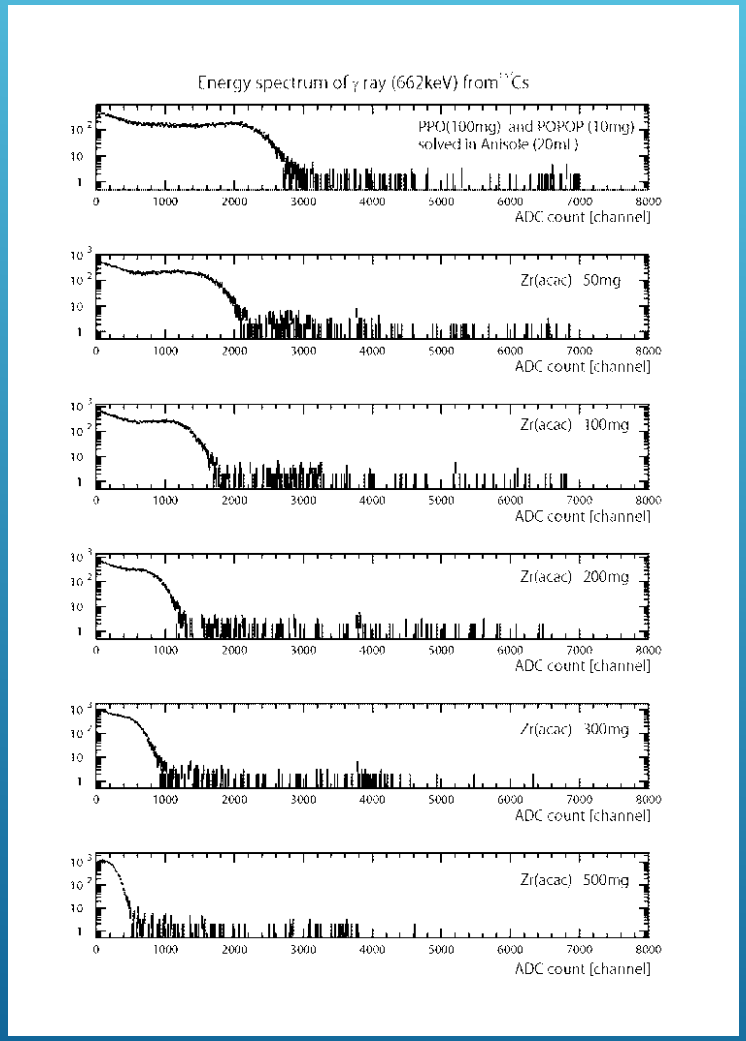


MW: 487.66



- 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.

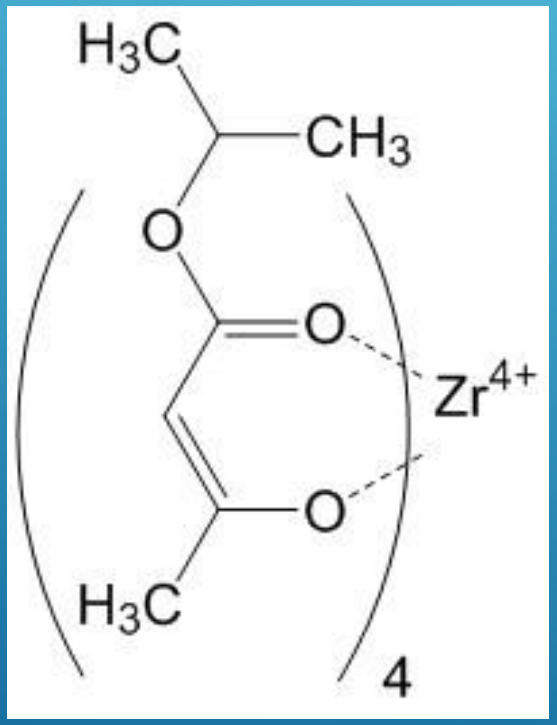
LIGHT YIELD AS A CONCENTRATION OF ZIRCONIUM ACETYLACETONATE



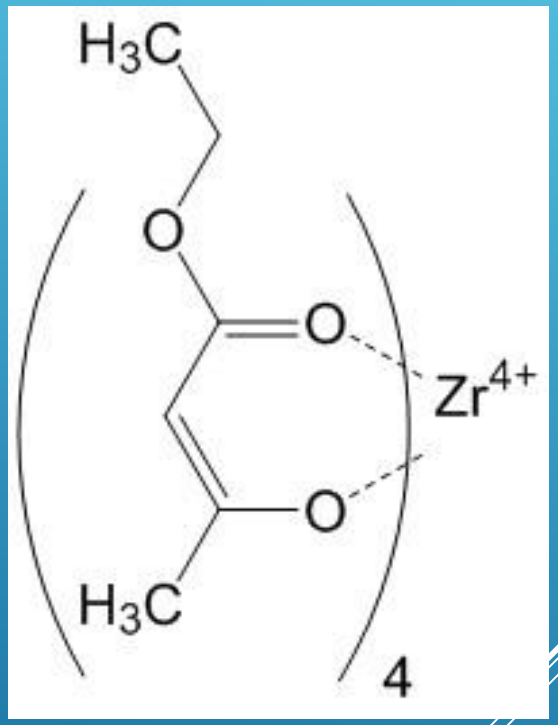
concentration of $Zr(acac)_4$	Observed channel	Expected channel
0 mg	2450	2450
50mg	1800	1997
100mg	1400	1687
200mg	950	1284
300mg	650	1038
500mg	300	750

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

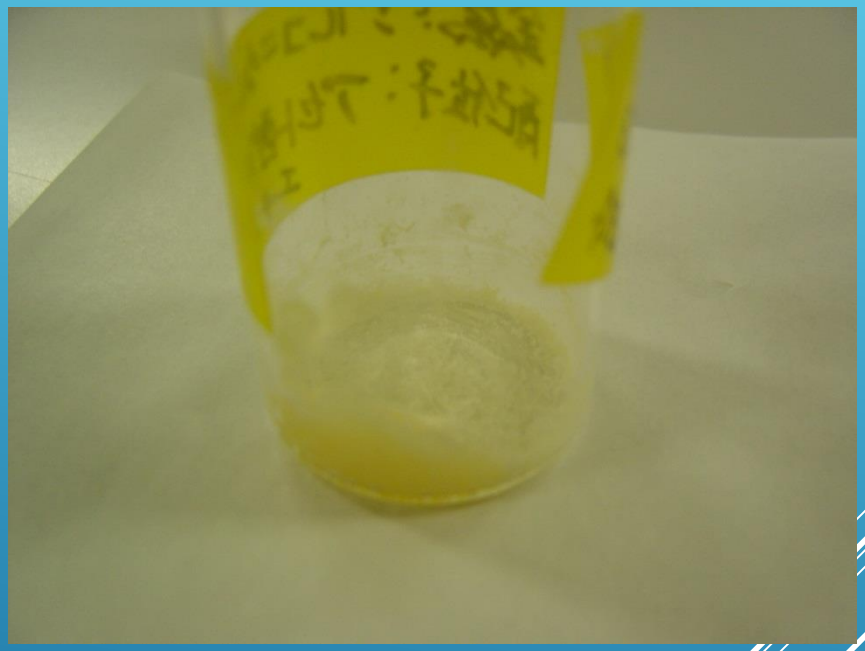


Expected shorter absorption wavelength

SYNTHESIZE OF ZIRCONIUM BETA-KETO ESTER COMPLEX

$Zr(iprac)_4$
state: powder

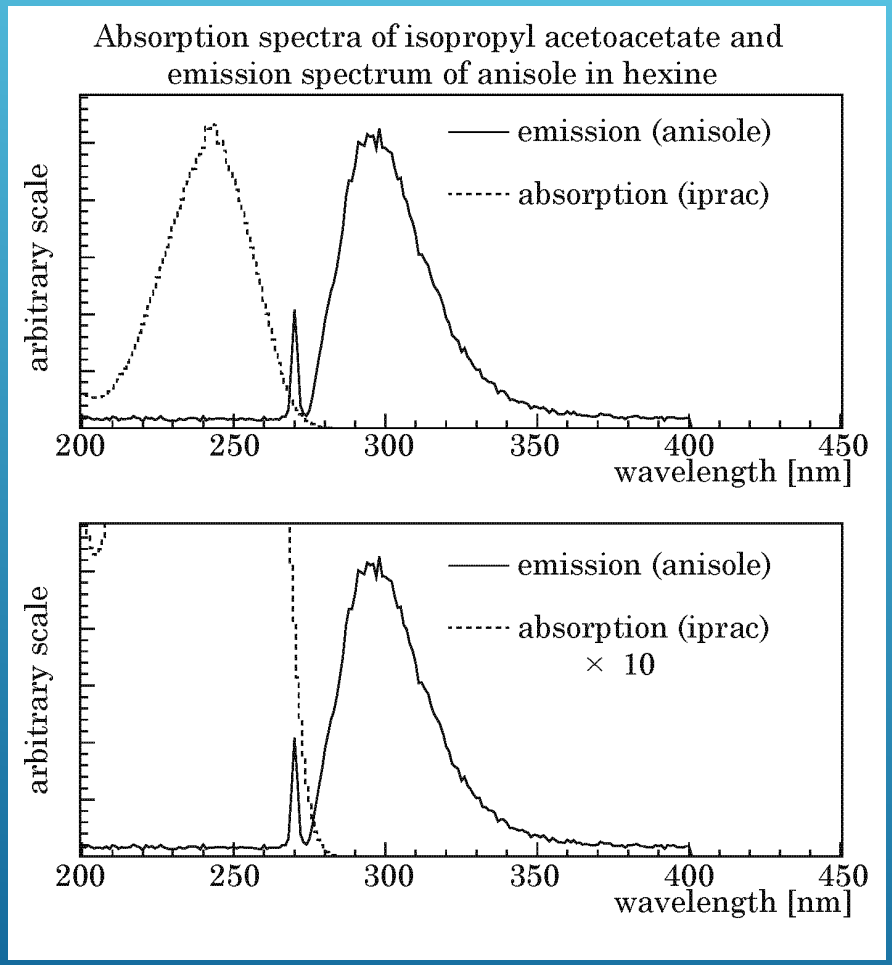
$Zr(etac)_4$
state : dry solid



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

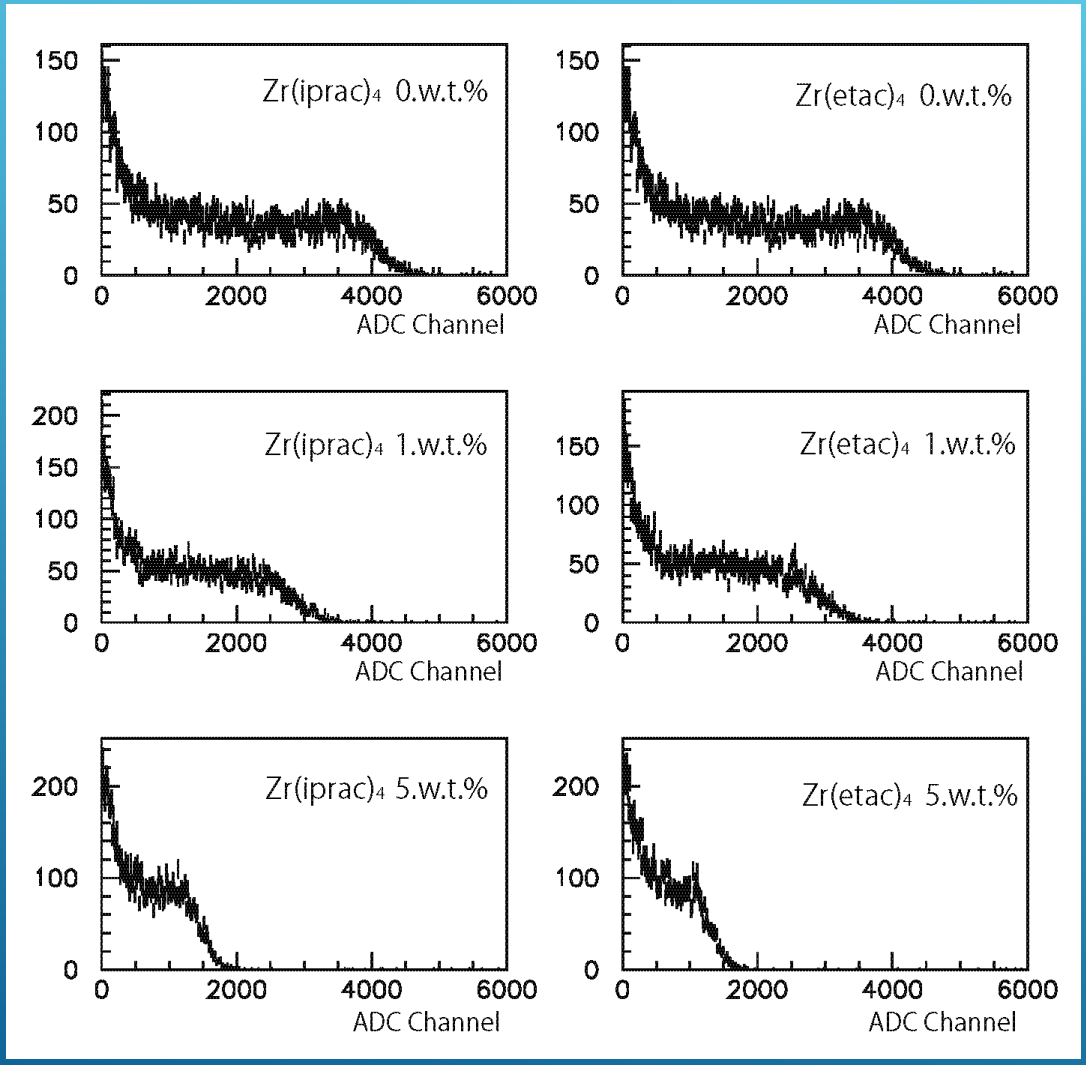
Solubility > 10 w.t.% for anisole

ABSORBANCE SPECTRA FOR ZIRCONIUM BETA-KETO ESTER COMPLEX



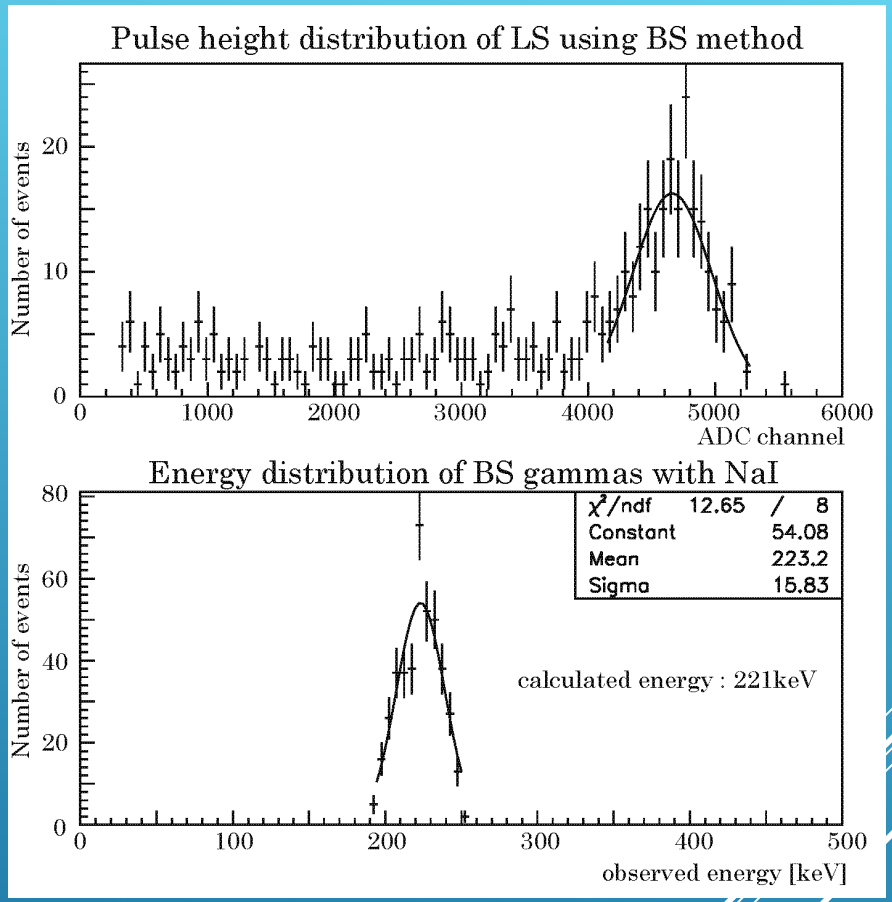
Absorption tail of β-keto ester ligands slightly overlapped with the region of the emission of anisole.

LIGHT YIELD OF LS CONTAINING ZIRCONIUM BETA-KETO ESTER COMPLEX



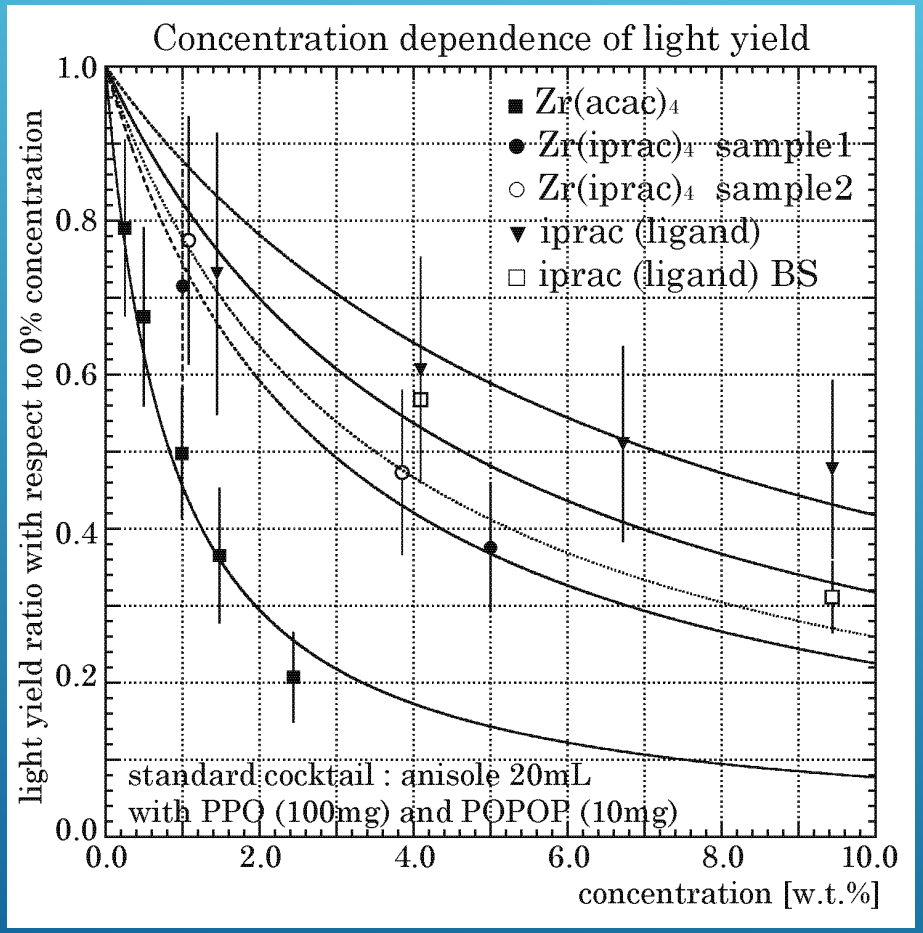
Light yield decreased as increasing the concentration of the complex even though small overlapping between the absorption of ligand and the emission of anisole .

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) . It is slightly smaller value than our goal...

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

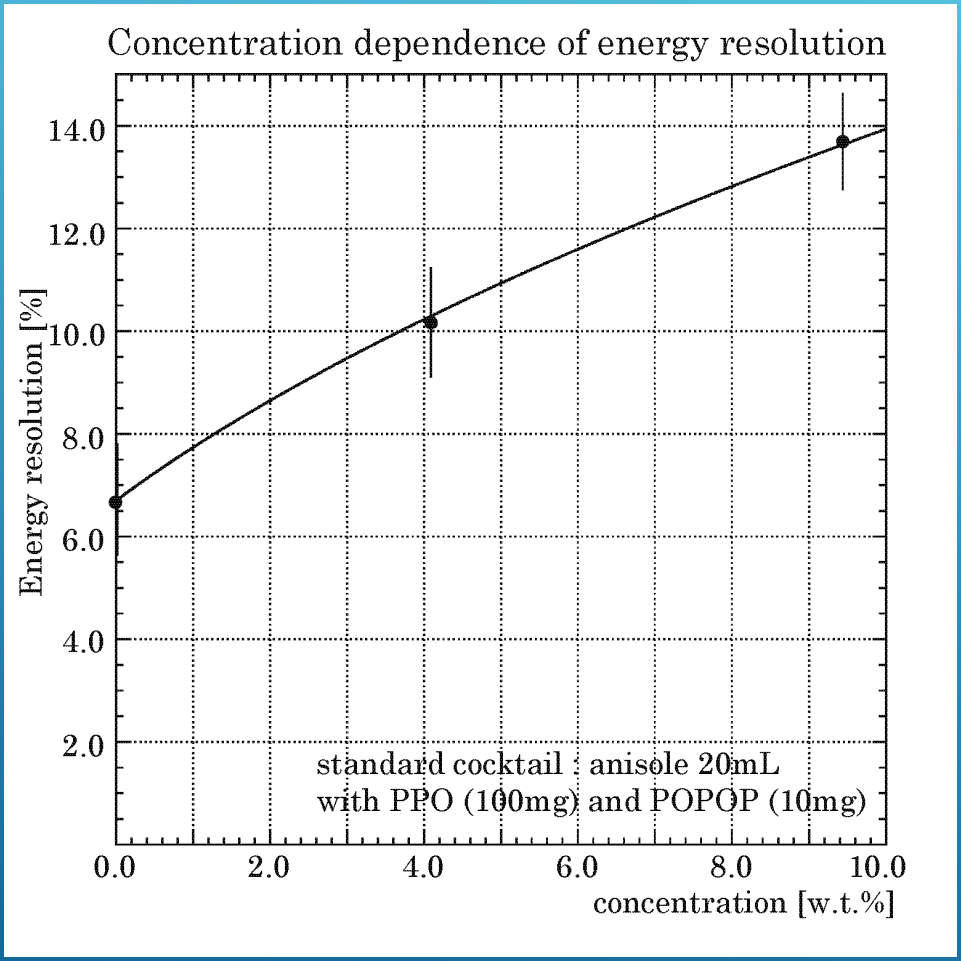
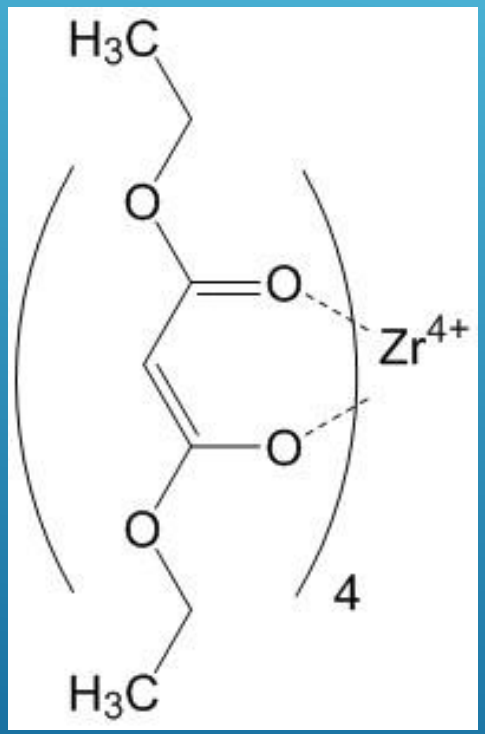


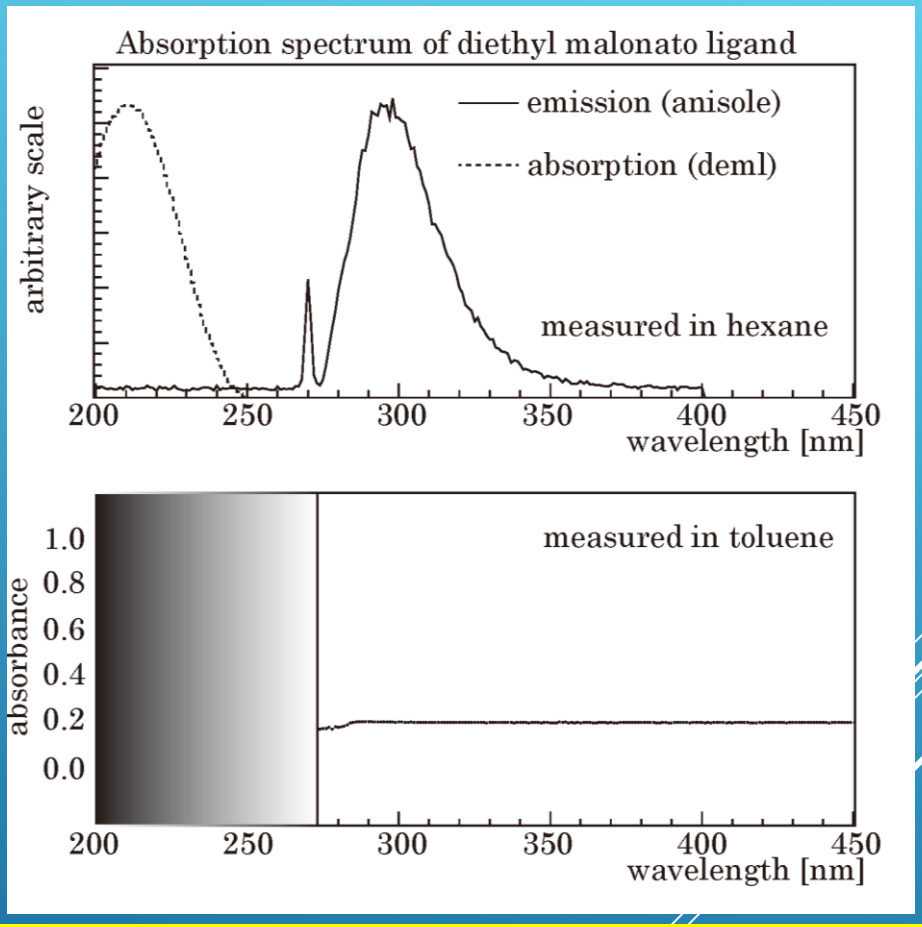
Photo coverage : ~8.5% (see explanation slide) Assuming 40% of photo coverage, the energy resolution will recover 6.5% @ 1MeV = 4.1% @ 2.5MeV for 10 w.t.% concentration.

They almost achieved to our initial goal!.

TETRAKIS (DIETHYL MALONATO) ZIRCONIUM AND ABSORBANCE SPECTRUM OF LIGAND

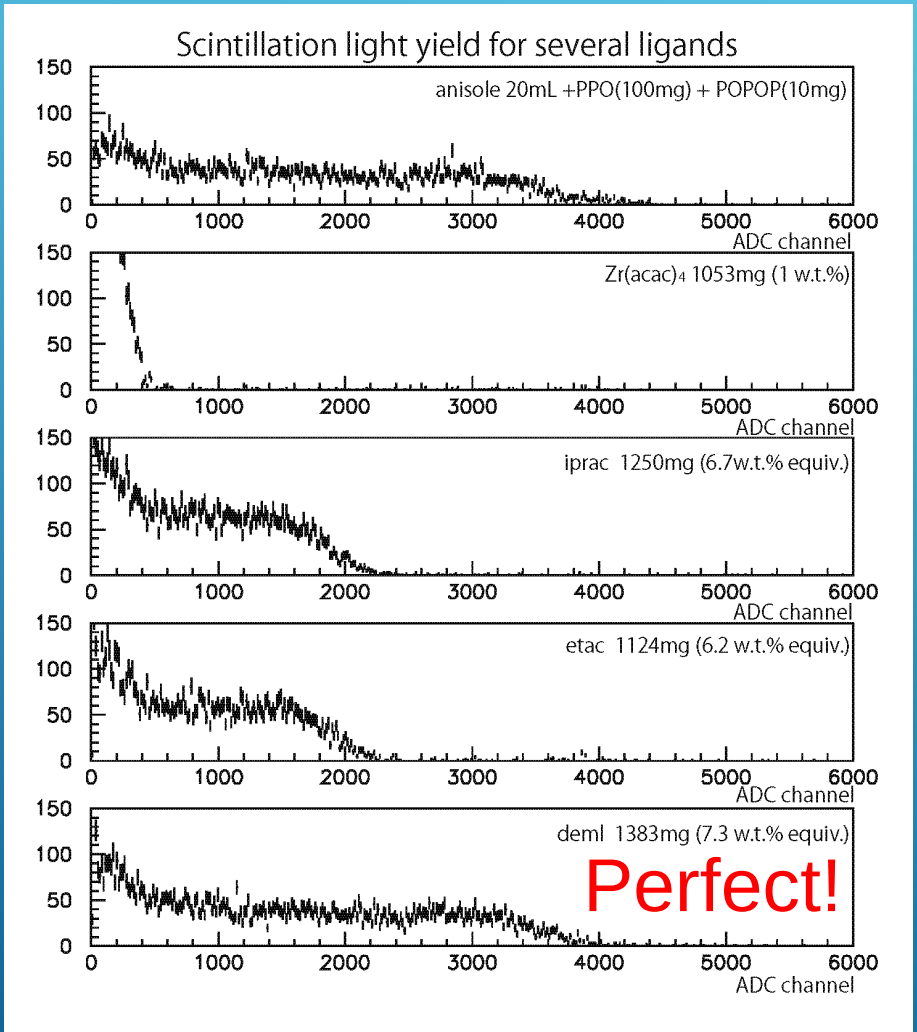
$Zr(CH_3CH_2OCOCHCOOCH_2CH_3)_4$
$= Zr(deml)_4$ mw : 727.87





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.

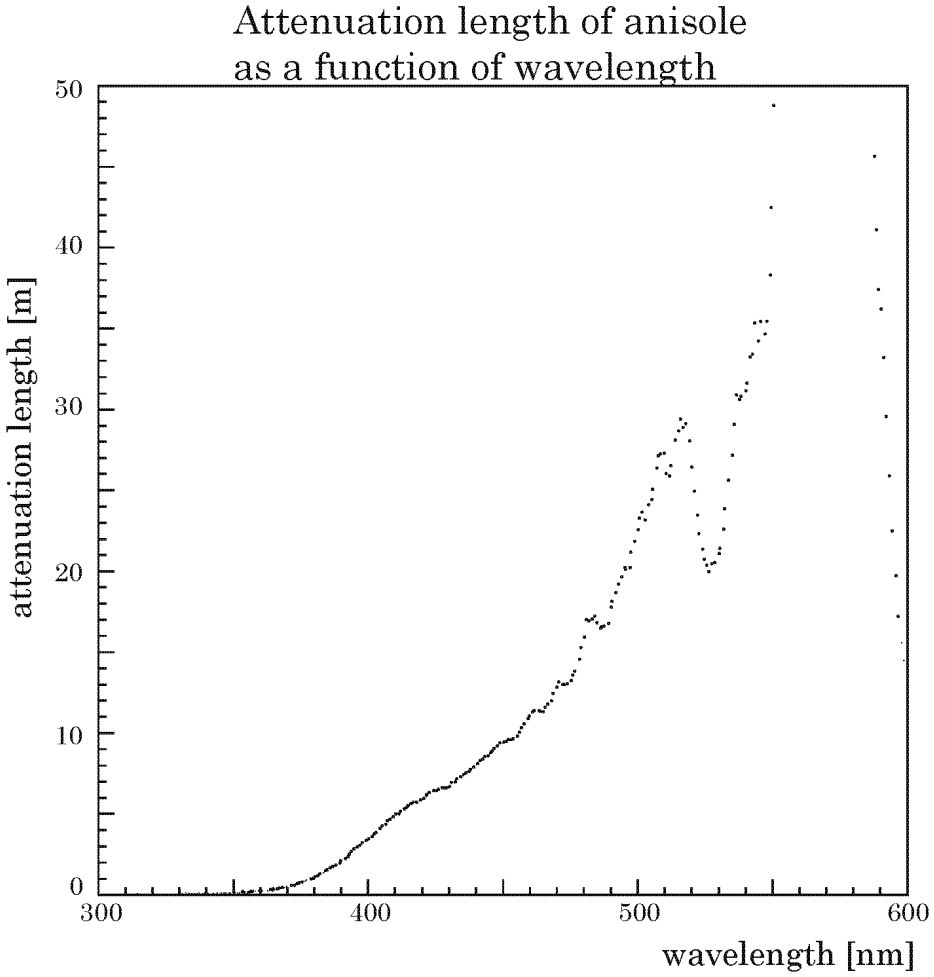
$Zr(deml)_4$ will be an ultimate complex if the solubility could be over 10w.t.% for anisole.

SUMMARY

- ZICOS uses liquid scintillator containing zirconium complex for neutrinoless double beta decay search
- Confirmed that the absorption peak moved shorter wavelength (275nm → 245nm) by introducing β-keto ester substituent groups.
- Succeeded anisole based liquid scintillator with **10 w.t.%** concentration of **$Zr(iprac)_4$** which 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 initial 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".

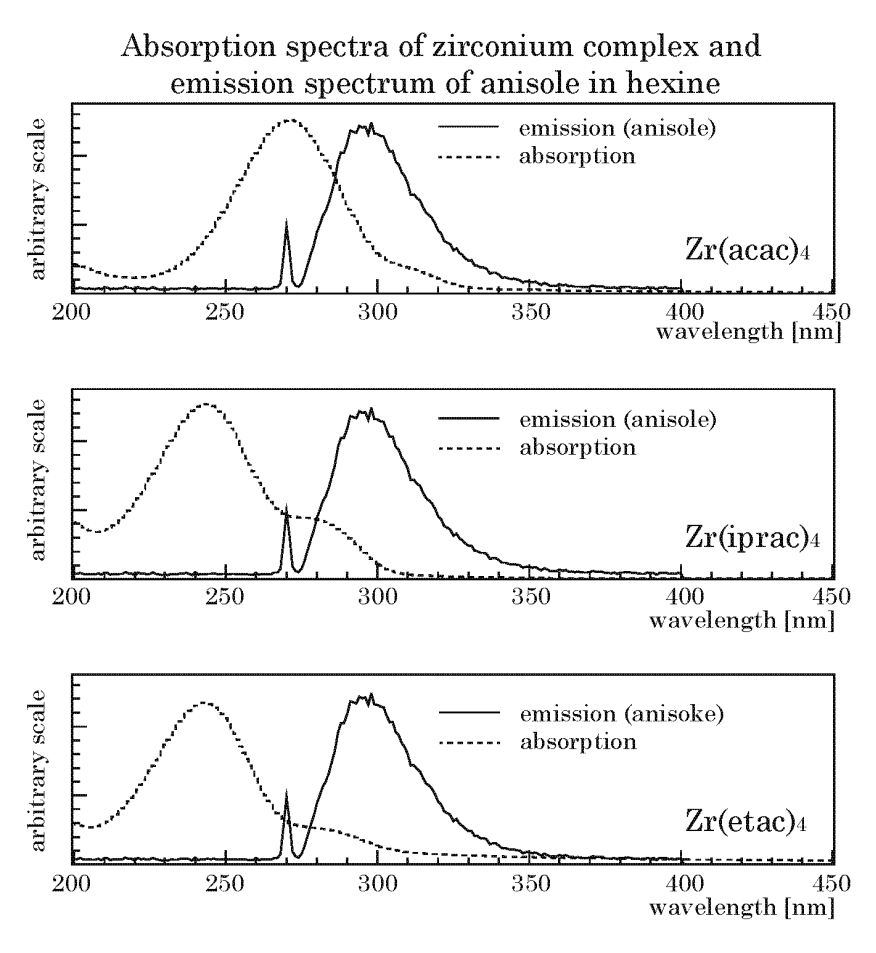
BACKUP

ATTENUATION LENGTH OF ANISOLE



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

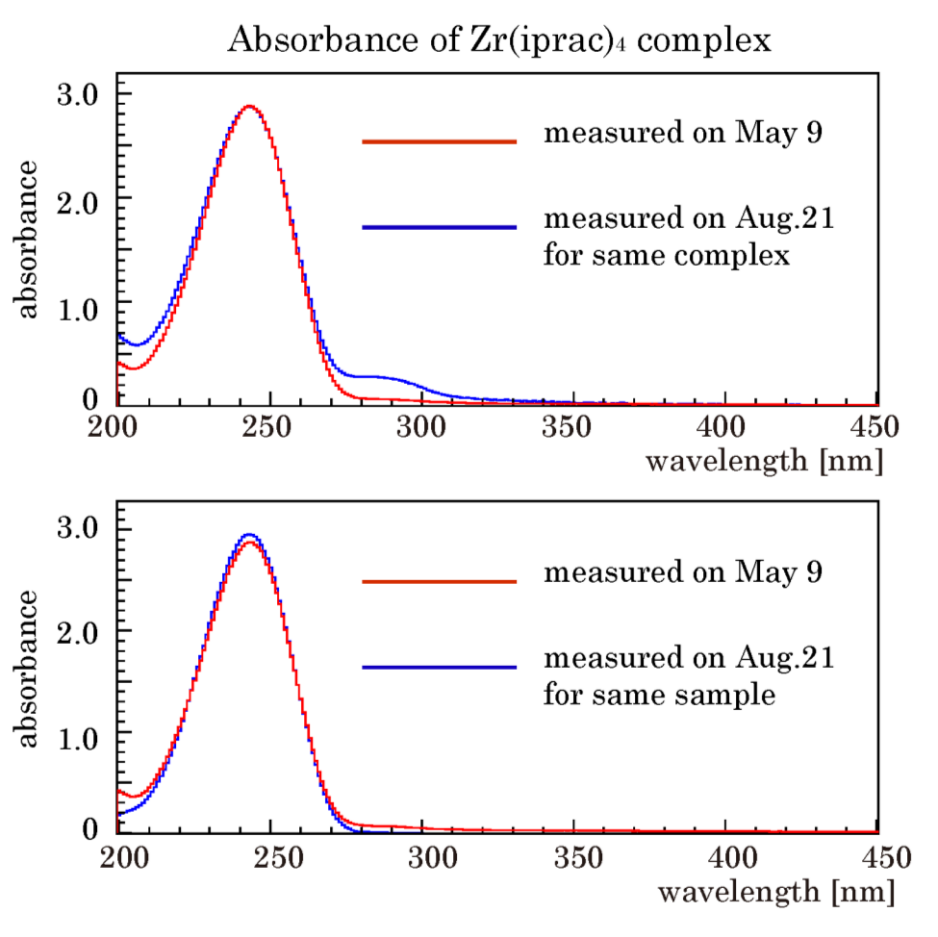
ABSORBANCE SPECTRA FOR ZIRCONIUM BETA-KETO ESTER COMPLEX



Absorption peaks of zirconium β-keto ester complex were found around at 245nm.

Also small bump was found around 280nm due to impurities which might be made by the vapor.

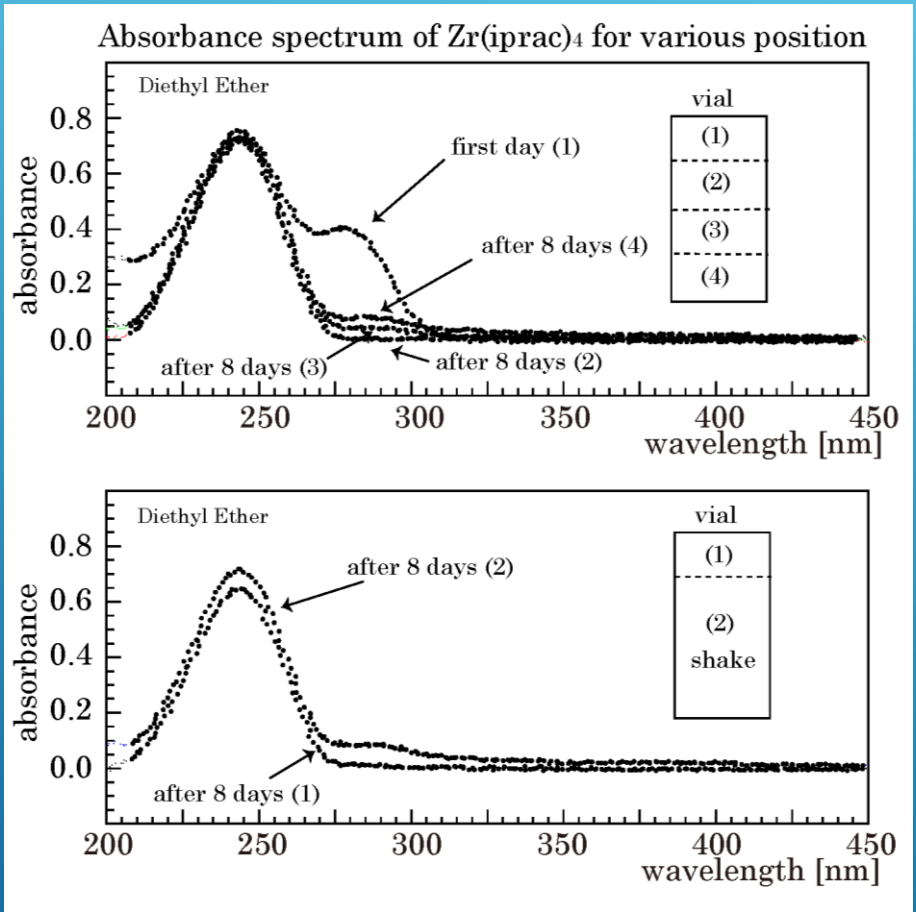
ABSORBANCE SPECTRA OF ZIRCONIUM BETA-KETO ESTER COMPLEX



No bump appeared for initial complex.

Small bump might be made by vapor in the air, because no bump was found for complex stored in solvent.

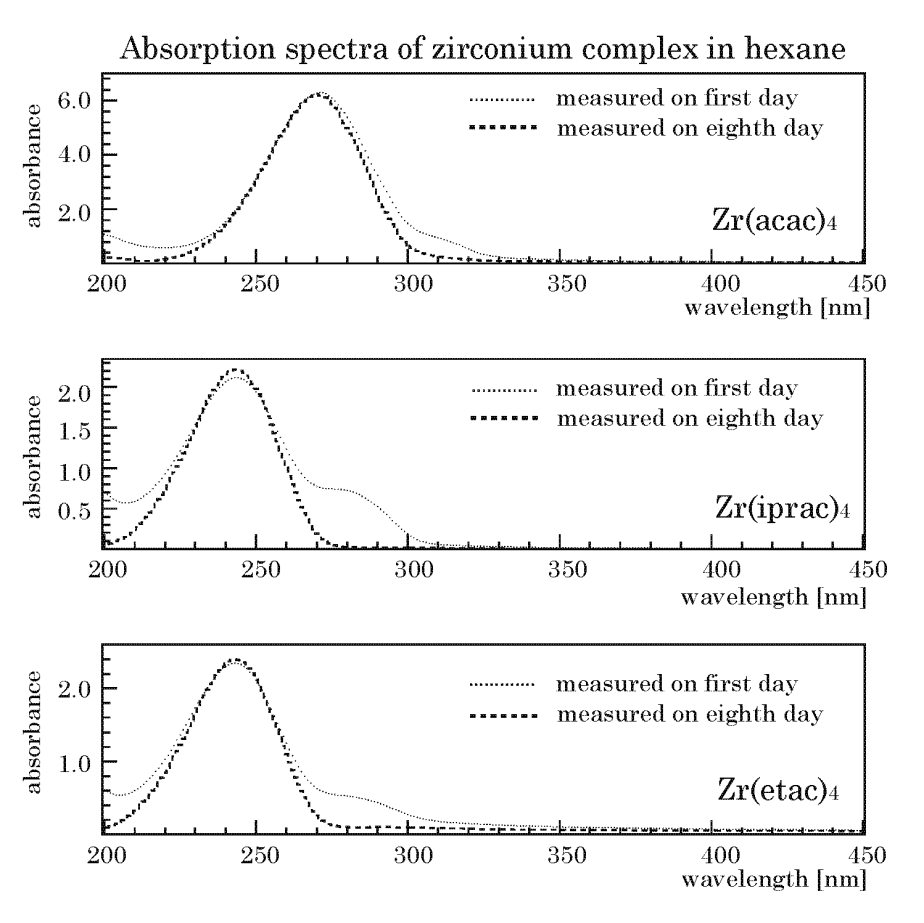
ABSORBANCE SPECTRA OF ZIRCONIUM BETA-KETO ESTER COMPLEX FOR VARIOS POSITION



Small bump disappeared after ~ 1 week.

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

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

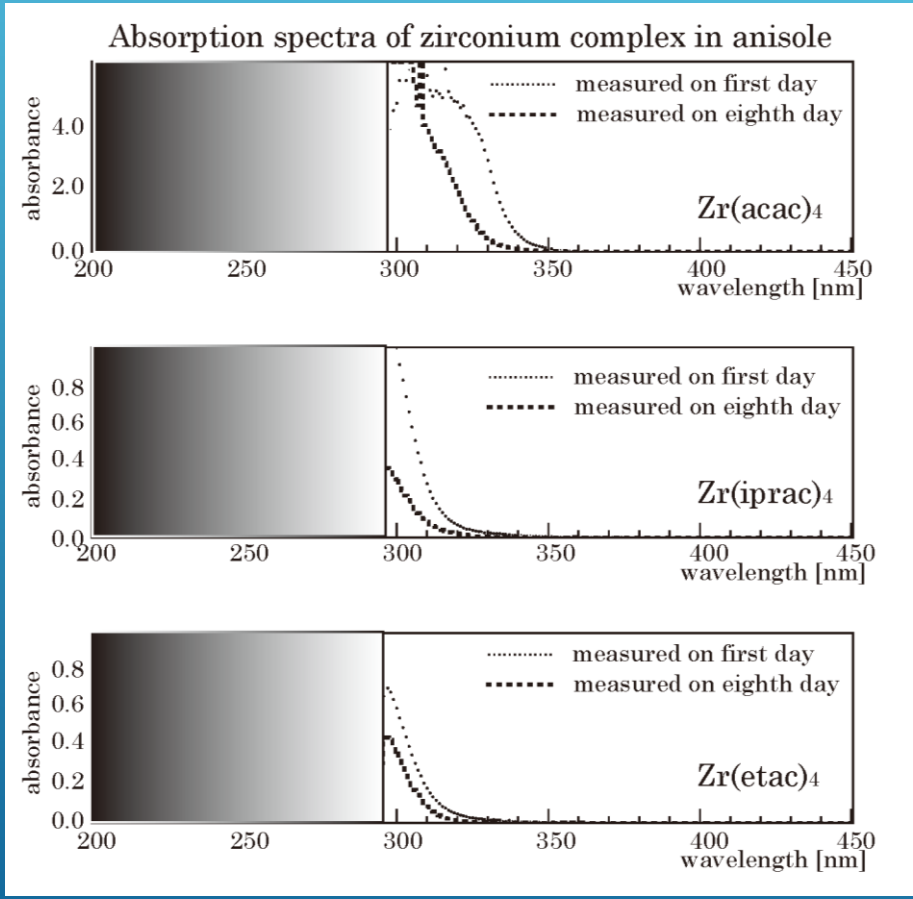


Small bump disappeared after ~ 1 week.

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

(see explanation slide)

ABSORBANCE SPECTRA FOR ZIRCONIUM BETA-KETO ESTER COMPLEX IN ANISOLE

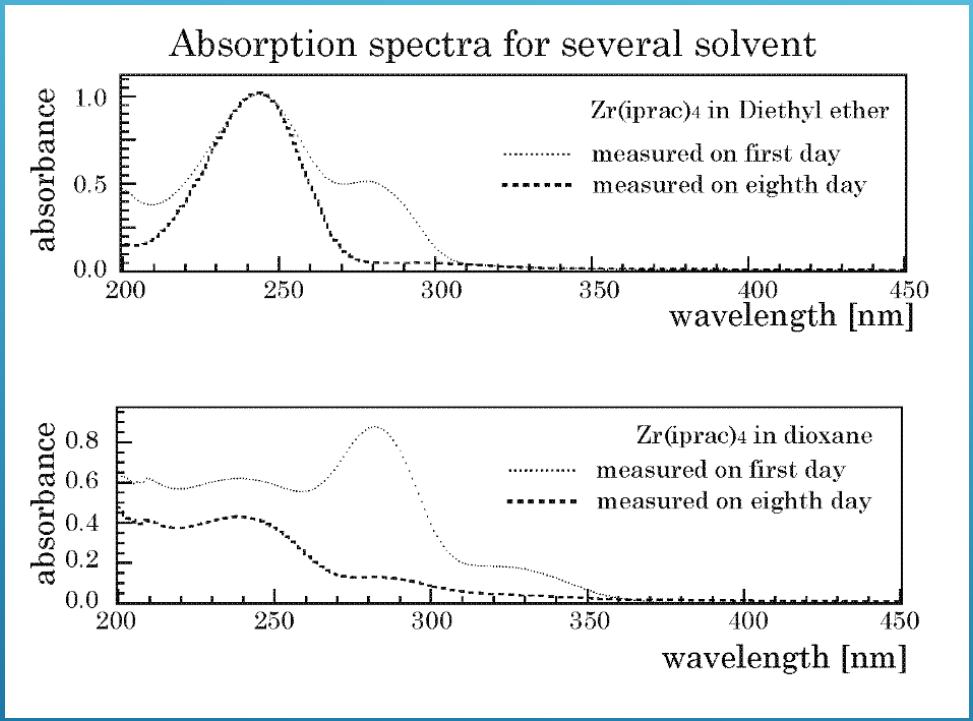


Small bump around 280nm in anisole did not disappear even after ~ 1 week.

Impurities might be dissolved in anisole…

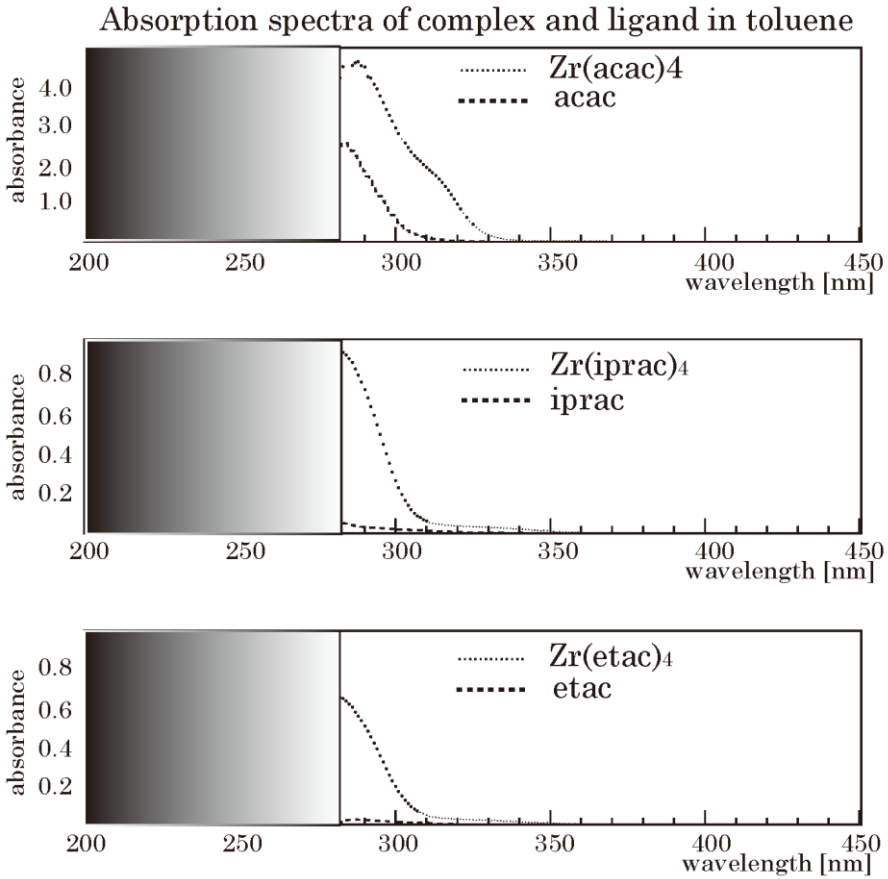
We have to purify the complex by using such as sublimation.

ABSORPTION SPECTRA FOR SEVERAL SOLVENT

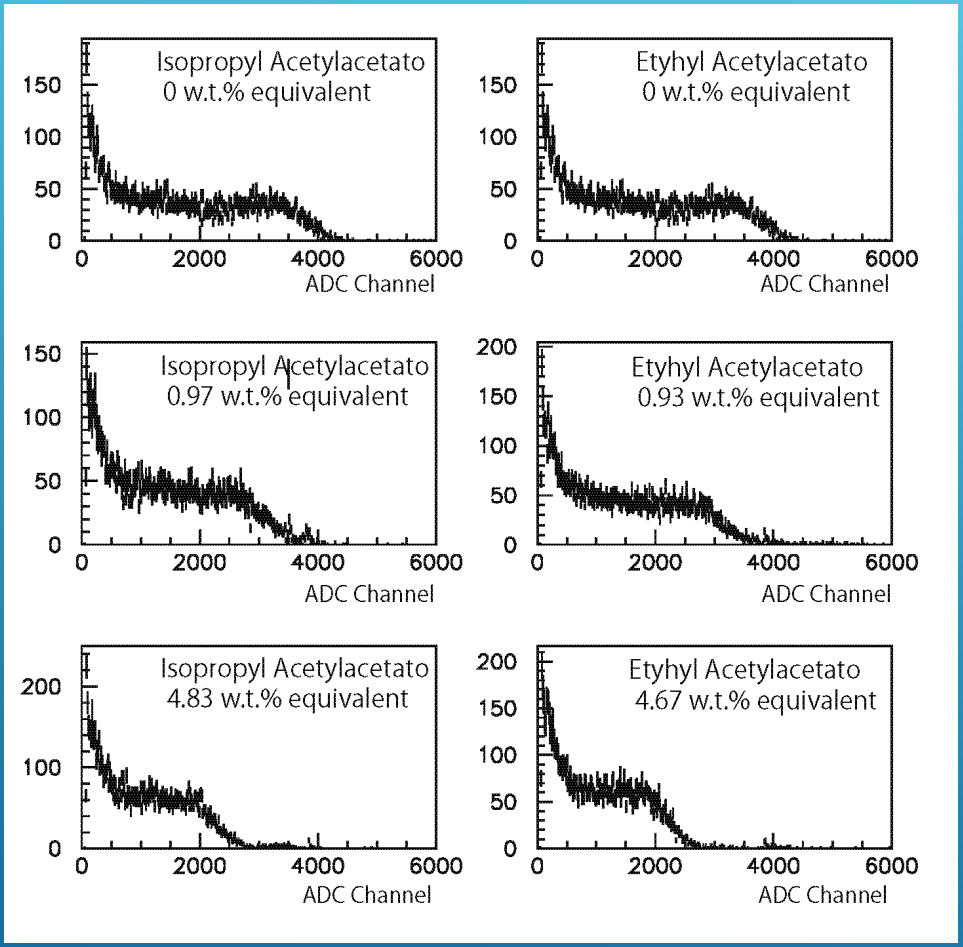


- Absorption spectra of Zr(iprac)4 changes.

ABSORPTION SPECTRA



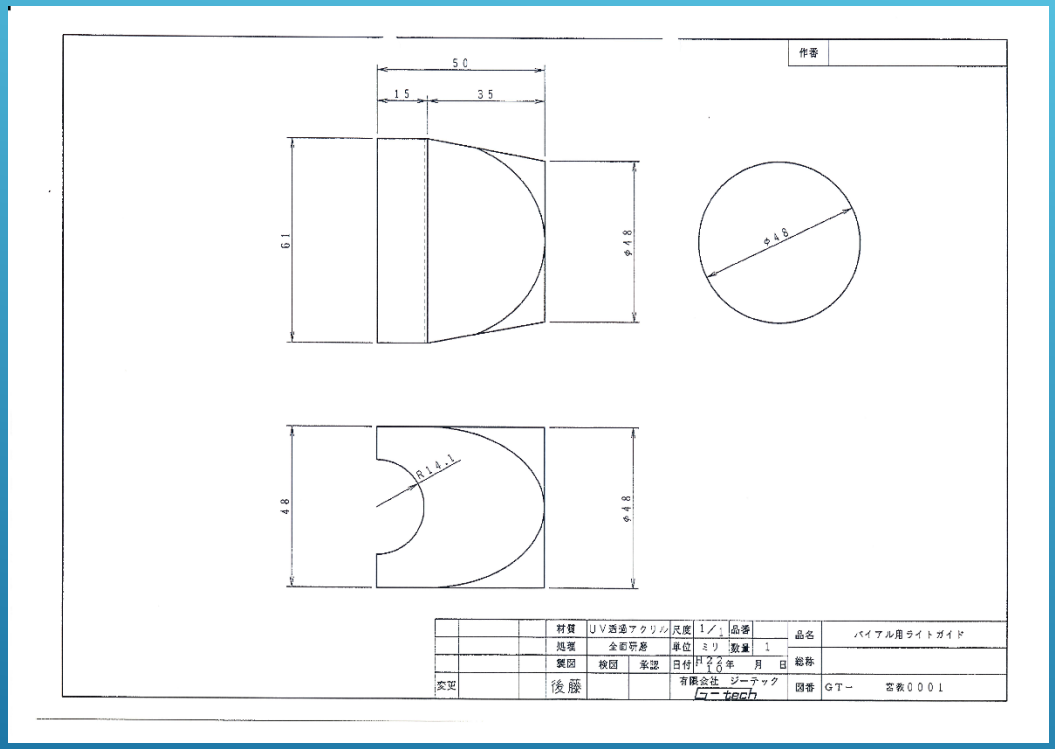
LIGHT YIELD OF LS CONTAINING BETA-KETO ESTER LIGAND



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

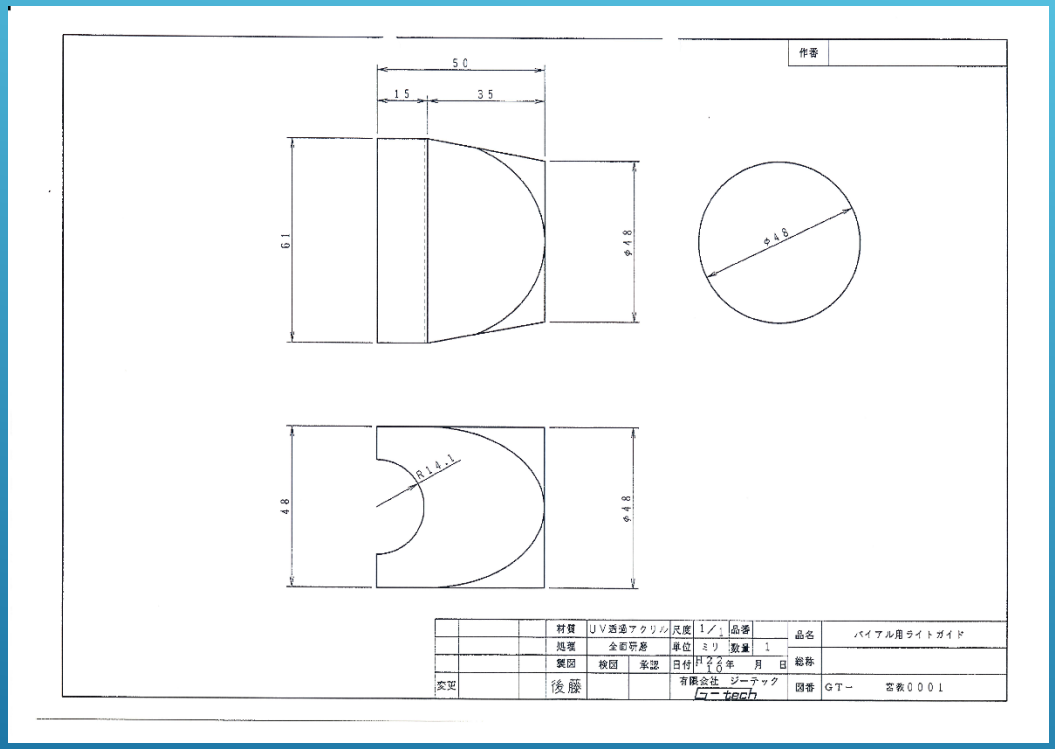
However, a quenching occurred due to the absorption tail.

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



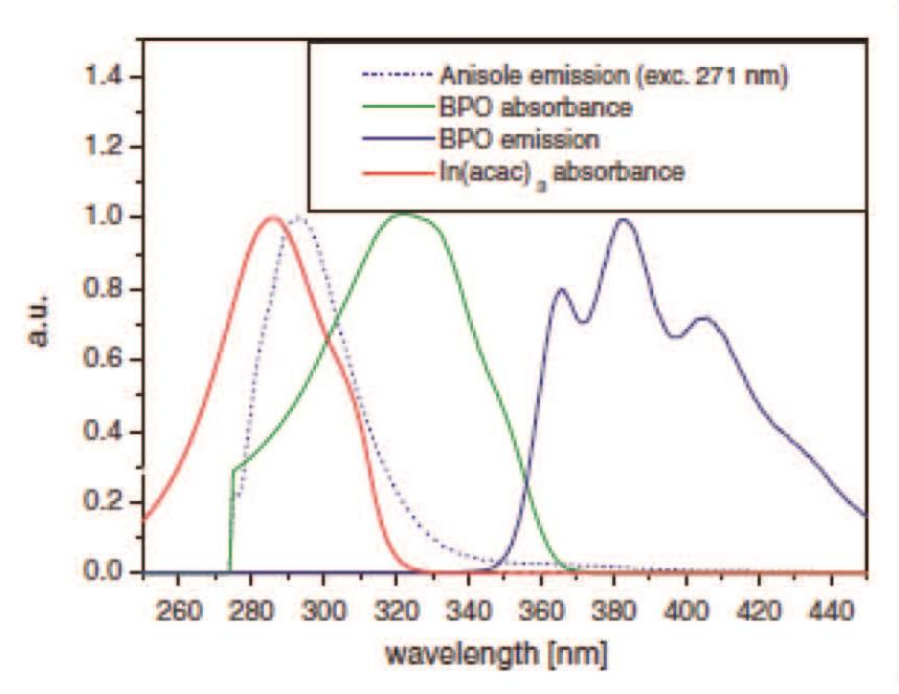
Scintillation light was guided to PMT via the acrylic light guide. From the computer simulation, the solid angle was obtained as 8.5% for 4π area.

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 complex molecular
σ_1: absorbance of PPO
σ_2 : absorbance of complex

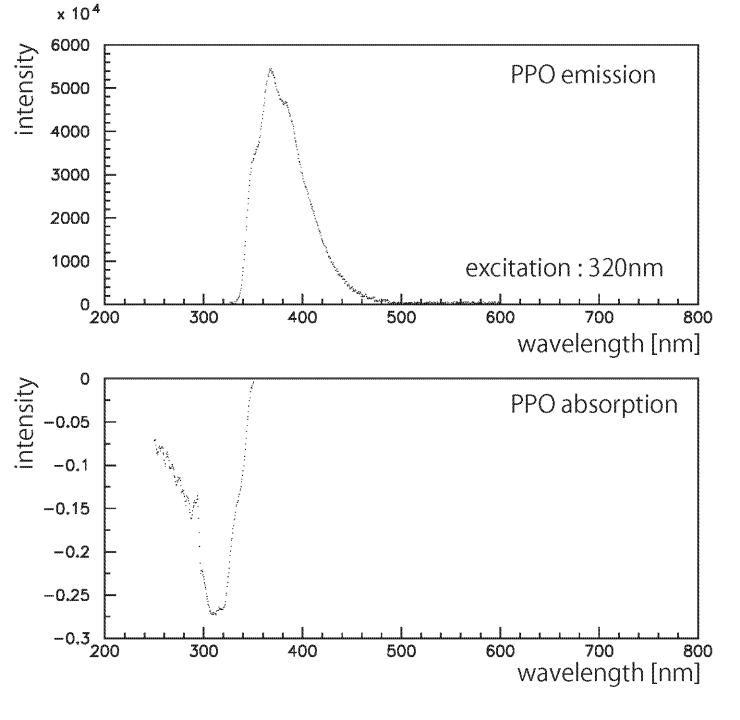
WHAT'S PROBLEM



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

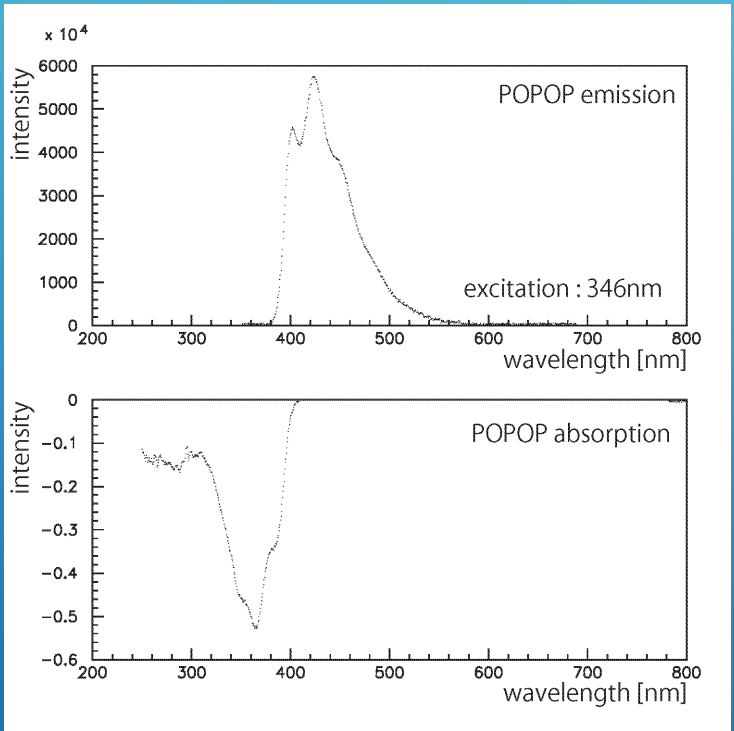
Same overlap between the emission and the absorption could be occurred even if different metal (Zr) was used.

PHOTO LUMINESCENCE AND ABSORPTION OF PPO



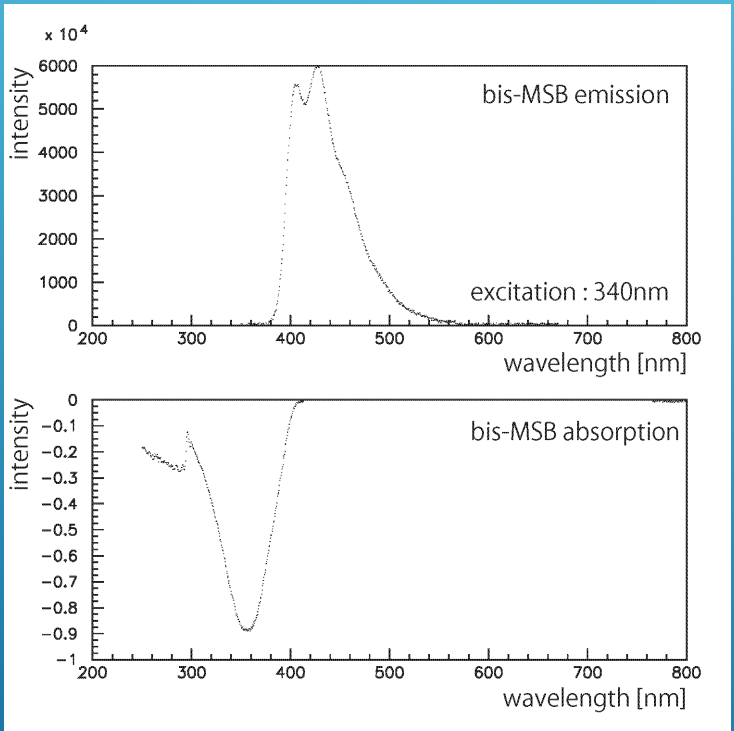
- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance device : HITACHI U-3000
  - Solvent : Benzonitrile (PhCN)
  - Concentration : 1.0×10^{-5} mol/L
  - 2,5-Diphenyloxazole
  - Molecular mass : 221.26
  - Max. emission wavelength : 368.0nm
  - Max. absorption wavelength : 309.7nm

PHOTO LUMINESCENCE AND ABSORPTION OF POPOP



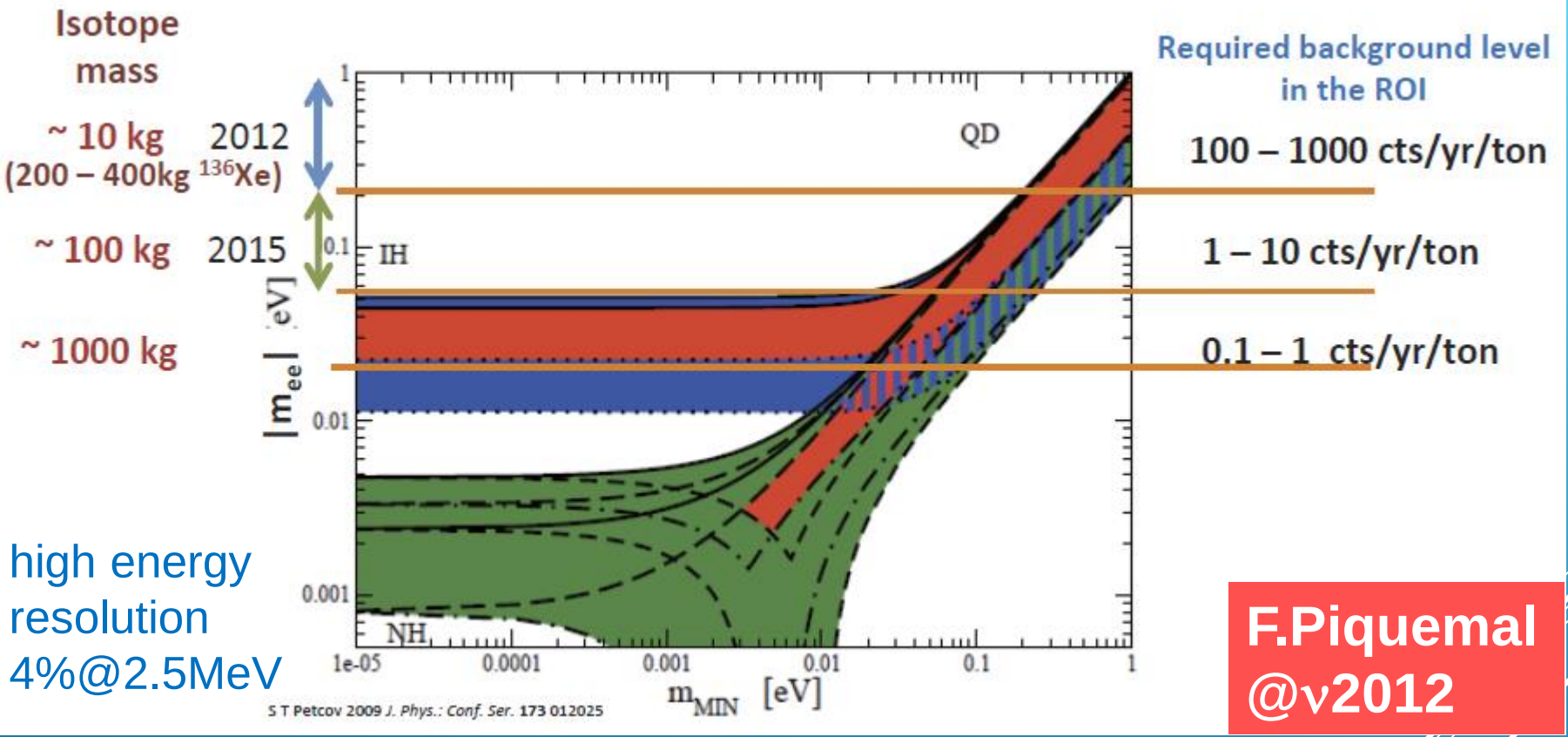
- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance device : HITACHI U-3000
  - Solvent : Benzonitrile (PhCN)
  - Concentration : 1.0×10^{-5} mol/L
  - 1,4-Bis(5-phenyloxazol-2-yl)benzene
  - Molecular mass : 364.40
  - Max. emission wavelength : 423.6nm
  - Max. absorption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

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