ジルコニウム錯体を用いた２重ベータ崩壊実験用液体シンチレータの開発

二重ベータ崩壊実験研究会（モンタナリゾート岩沼）
2010年12月17日

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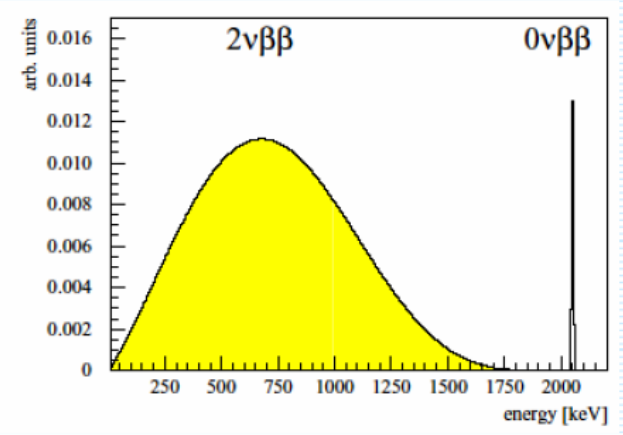
Motivation

- Neutrinoless double beta decay
  - Lifetime and neutrino mass

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

  - Energy spectrum and lifetime measurement



$$T_{1/2} \sim a\sqrt{\frac{M\,t}{\Delta E\,B}}$$

a : *isotopical abundance*
M : active mass
t : measuring time
ΔE: energy resolution
B : background count rate

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

For $0\nu\beta\beta$ experiment

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

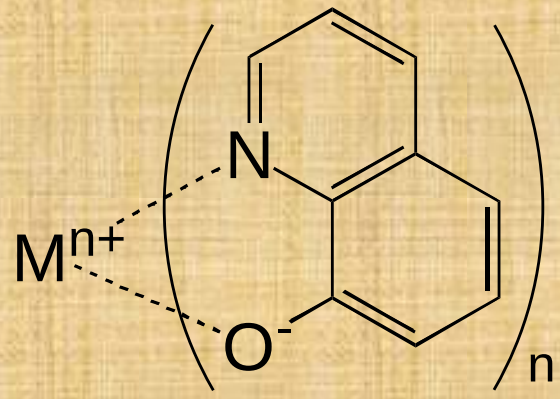
Transition	$Q_{\beta\beta}$ (keV)	Abundance (%) ($^{232}Th = 100$)
$^{110}Pd \rightarrow ^{110}Cd$	2013	12
$^{76}Ge \rightarrow ^{76}Se$	2040	8
$^{124}Sn \rightarrow ^{124}Te$	2288	6
$^{136}Xe \rightarrow ^{136}Ba$	2479	9
$^{130}Te \rightarrow ^{130}Xe$	2533	34
$^{116}Cd \rightarrow ^{116}Sn$	2802	7
$^{82}Se \rightarrow ^{82}Kr$	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

- Above ^{208}Tl γ line (2.614MeV)
- Formed metal complex and solved in organic solvent ⇒ Zirconium (Zr)

Metal complex for liquid scintillator

- metal complex

  tris(8-quinolinolate) metal complex (MQ_n)

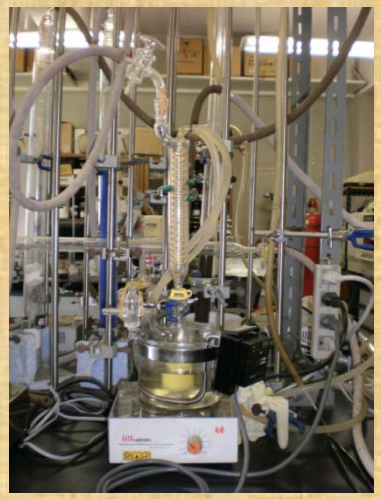


M = In, n = 3; M = Zr, n = 4

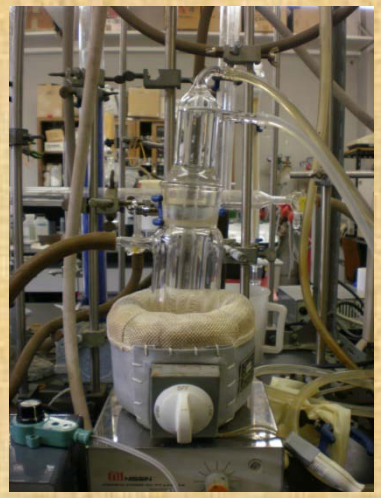
- AlQ_3 has been established as organic Electro Luminescence material (@530nm)
- InQ_3 and ZrQ_4 should also have same property of luminescence with almost same wavelength

Desired performcance
Light yield : 60% of BC505
Energy resolution : 4%@3MeV

Synthesis of ZrQ_4 and sublimation








InQ_3
primary yield 100%
sublimation 77%

ZrQ_4
primary yield 96%
sublimation 70%

Solution InQ_3 and ZrQ_4 in organic solvent

- InQ_3 and ZrQ_4 dissolved in Benzonitrile (PhCN) with ~2%
  - Benzonitrile (PhCN: C_6H_5CN)

    density : 1.0g/mL

    flash point : 75℃

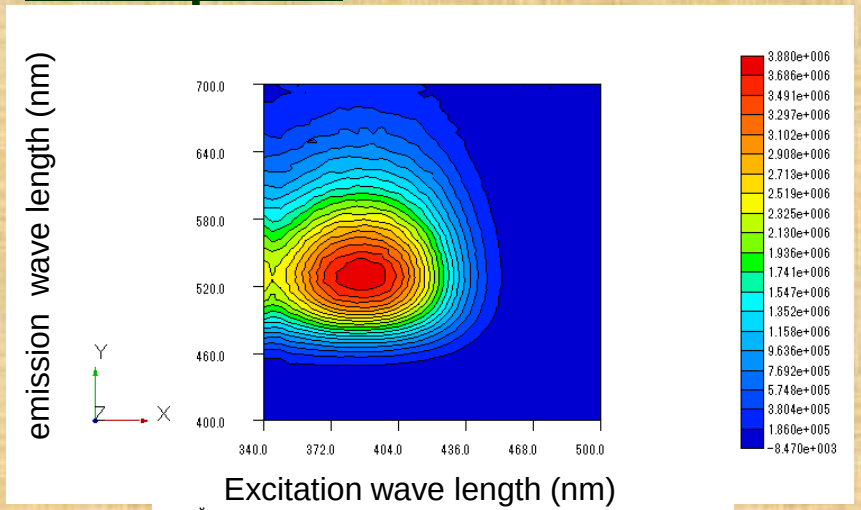
    photon emission : 291nm@maximum

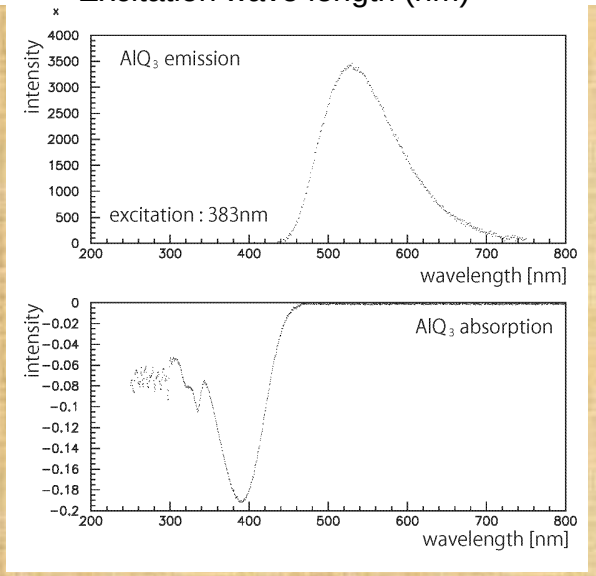
    attenuation length : 66cm (@0.5wt%)
  - Liquid scintillator cocktail

    PhCN+PPO(100mg)+POPOP(10mg)

    PhCN+PPO(100mg)+bis-MSB(10mg)

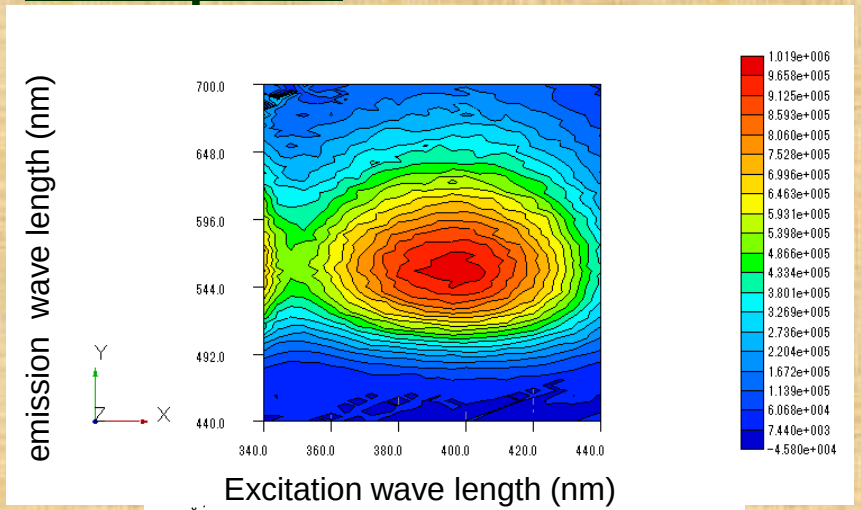
Photo Luminescence and absorption of AlQ_3 complex

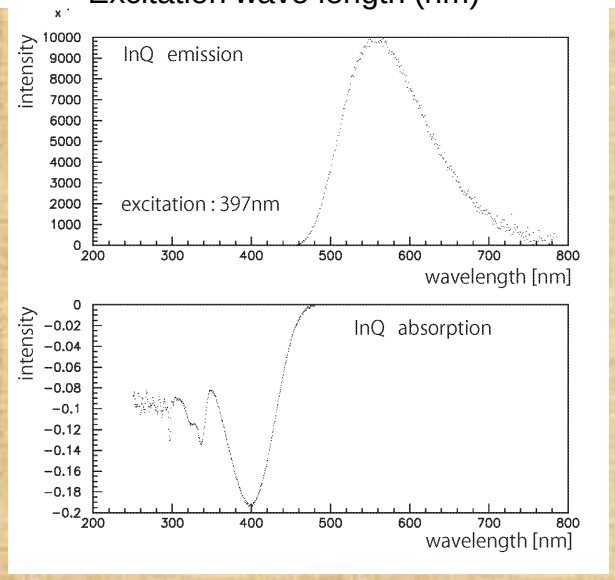




- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance devie : HITACHI U-3000
  - Solvent : Benzonitrile (PhCN)
  - Concentration : 3.0×10^{-5} mol/L
  - Molecular mass : 459.44
  - Max. emission wavelength : 529.9nm
  - Max. absorption wavelength : 389.6nm

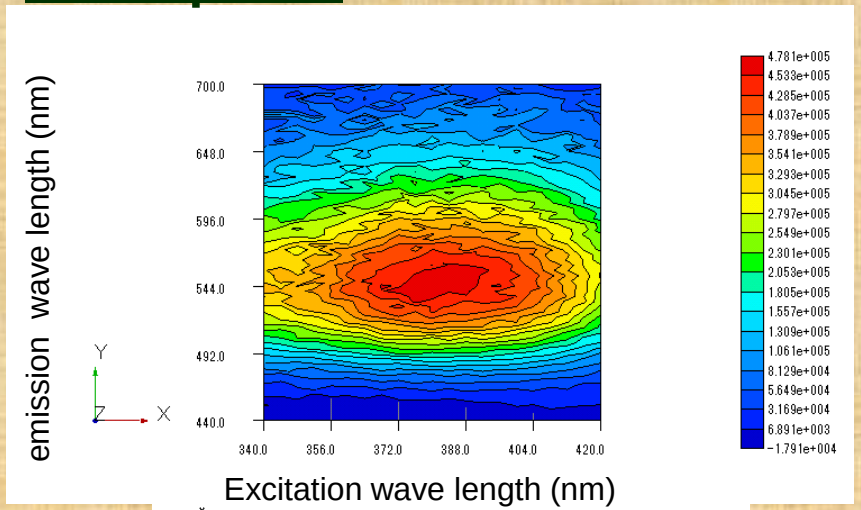
Photo Luminescence and absorption of InQ_3 complex

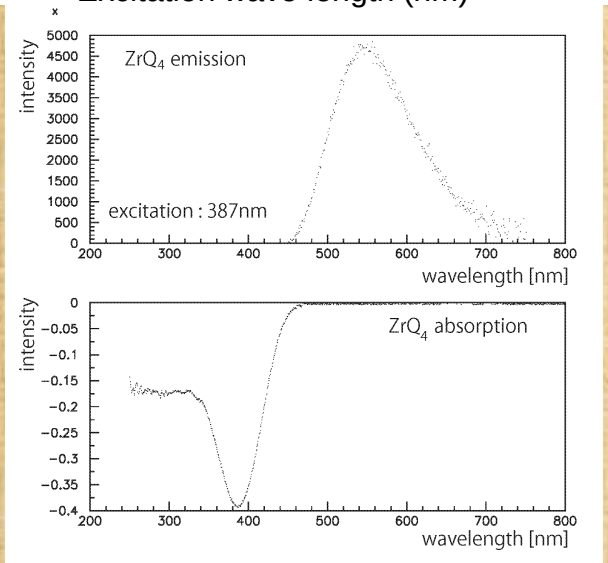




- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance devie : HITACHI U-3000
  - Solvent : Benzonitrile (PhCN)
  - Concentration : 3.0×10^{-5} mol/L
  - Molecular mass : 547.28
  - Max. emission wavelength : 559.3nm
  - Max. absorption wavelength : 394.7nm

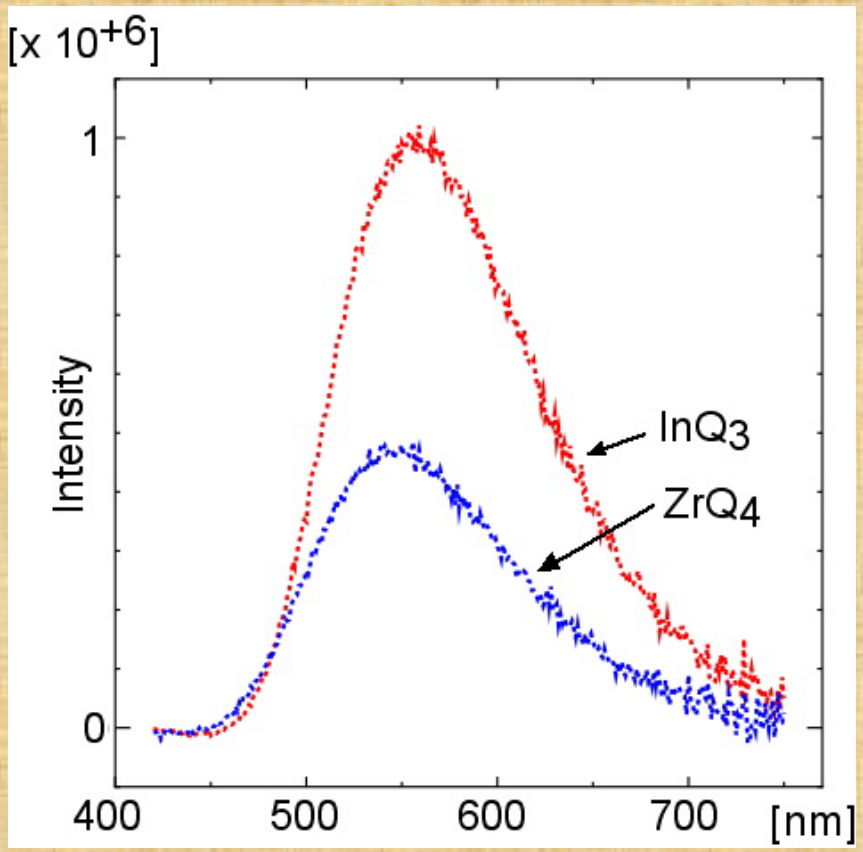
Photo Luminescence and absorption of ZrQ_4 complex





- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance devie : HITACHI U-3000
  - Solvent : Benzonitrile (PhCN)
  - Concentration : 3.0×10^{-5} mol/L
  - Molecular mass : 667.84
  - Max. emission wavelength : 548.0nm
  - Max. absorption wavelength : 383.3nm

Quantum yield of InQ_3 and ZrQ_4 for photo luminescence



- Quantum yield
  - Fluorescence intensity (area of wavenumber spectra) of Quinine as standard (Ir)
  - Same intensity using corrected spectrum for InQ_3/ZrQ_4 was calculated (Is)
  - Absorbance of Quinine (Ar:0.34) and InQ_3/ZrQ_4 (As:0.19/0.39)
  - Quantum Yield is defined by

$$\Phi s = \Phi r(Is/Ir)(Ar/As)$$

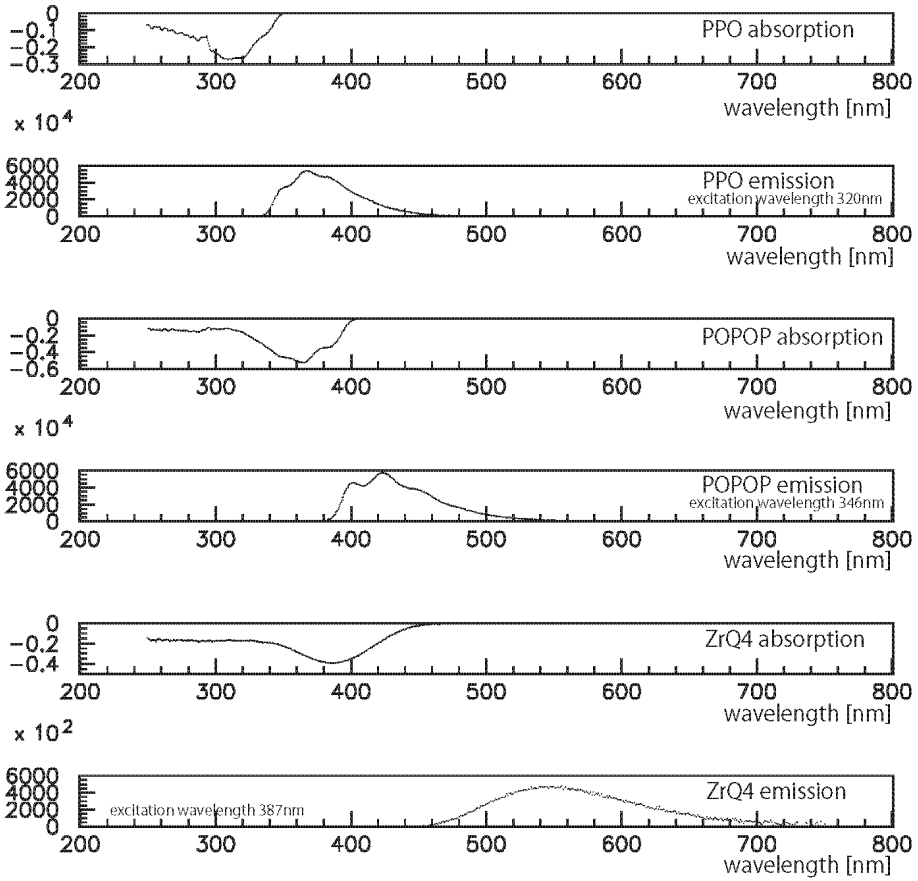
$\Phi s(InQ_3) = 0.050$ [Is/Ir = 0.051]

$\Phi s(ZrQ_4) = 0.011$ [Is/Ir = 0.023]

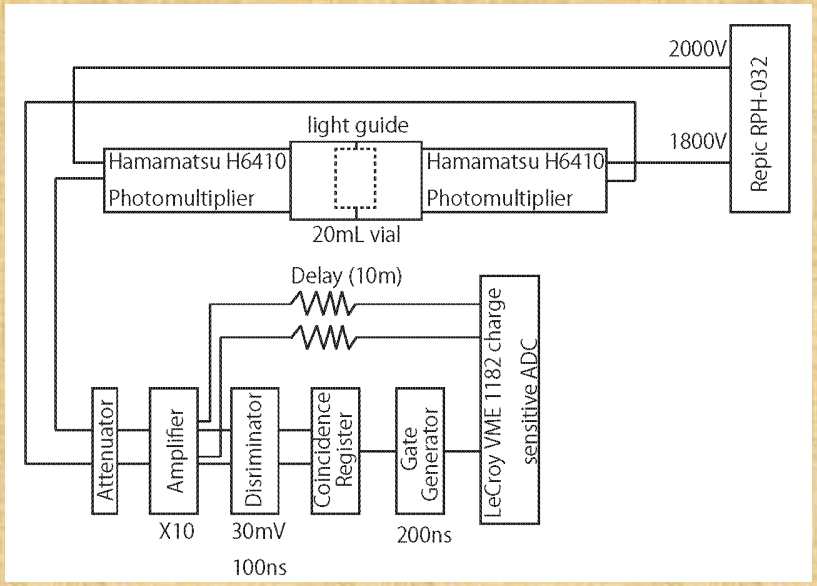
cf. $\Phi s(AlQ_3)=0.17$

Energy transfer via photon radiation

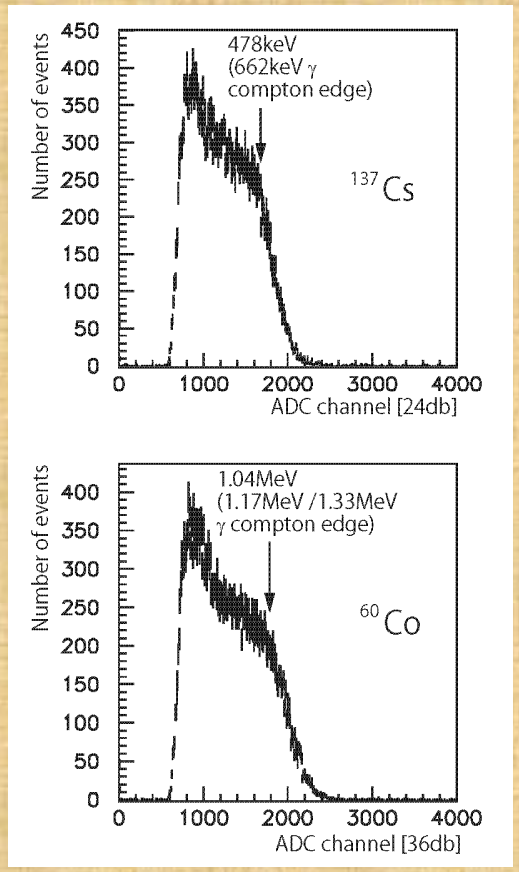
- PPO→POPOP→ZrQ_4



DAQ setup



BC505 (standard scinti.)



1040keV :
1630ch@30db

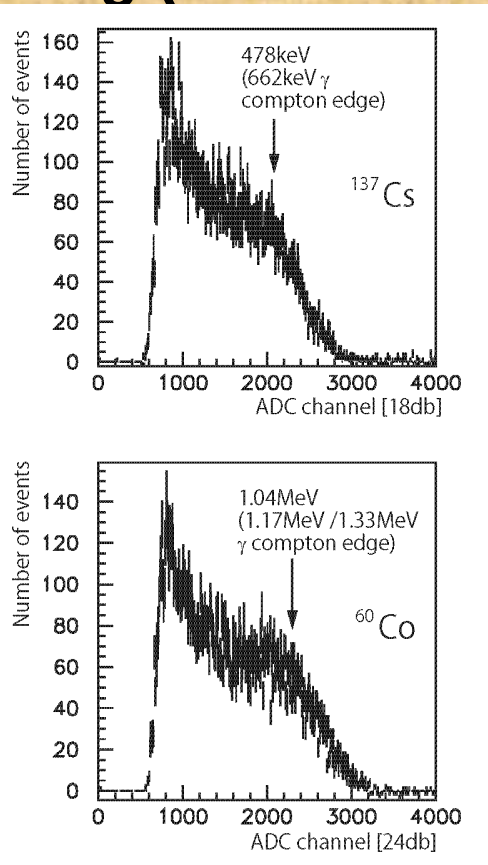
52160ch@0db
(QE : 0.25)

208640ch@0db

478keV:
1575ch@24db
expected :
1498ch

Comparison of light yield of PhCN scintillator

PhCN/PPO100mg/POPOP 10mg (PhCN-POPOP)



1040keV :
2075ch@24db

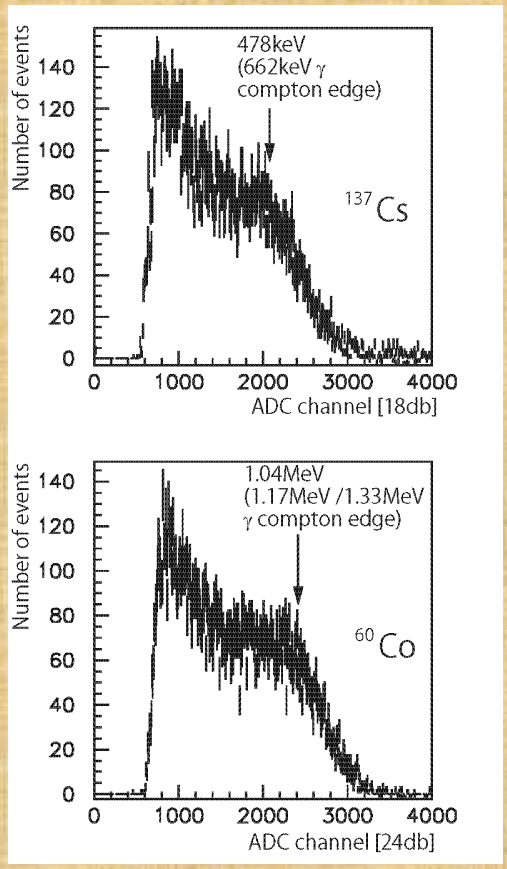
33200ch@0db
(QE : 0.25)

132800ch@0db

Light Yield
toBC505: 63%

478keV:
1875ch@18db
expected :
1907ch

PhCN/PPO100mg/bis-MSB 10mg (PhCN-bisMSB)



1040keV :
2255ch@24db

36080ch@0db
(QE : 0.25)

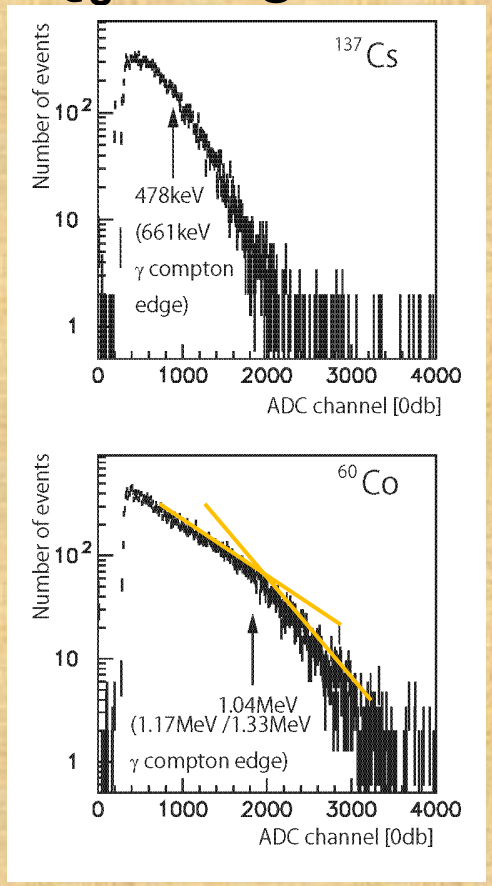
144320ch@0db

Light Yield
toBC505: 69%

478keV:
2025ch@18db
expected :
2072ch

Response for γ–ray from radioactive source (1) : Quantum Yield and Light Yield

InQ_3 50mg in PhCN-POPOP



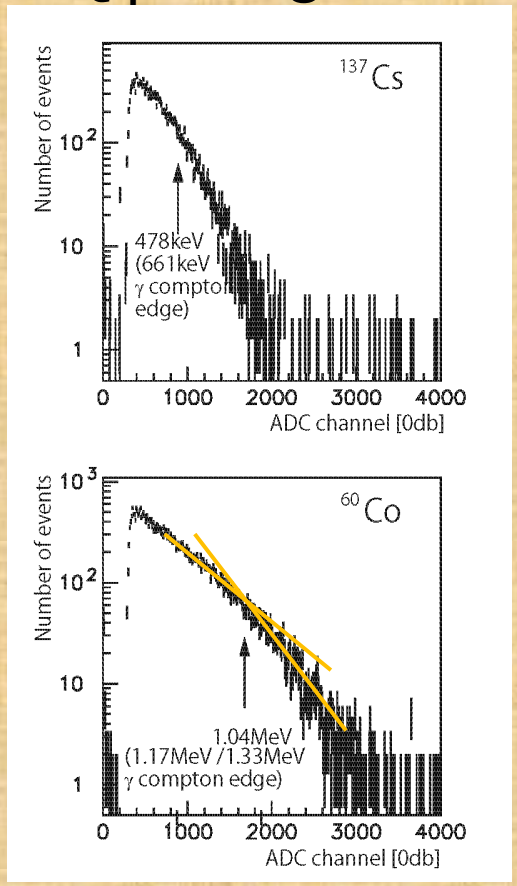
1040keV :
1925ch@0db
(QE : 0.093)

20699ch@0db

Quantum Yield :
20699/132800
=**15.6%(5.0%)**

Light Yield to BC505:
20699/208640
=9.9%

ZrQ_4 50mg in PhCN-POPOP



1040keV :
1525ch@0db
(QE : 0.10)

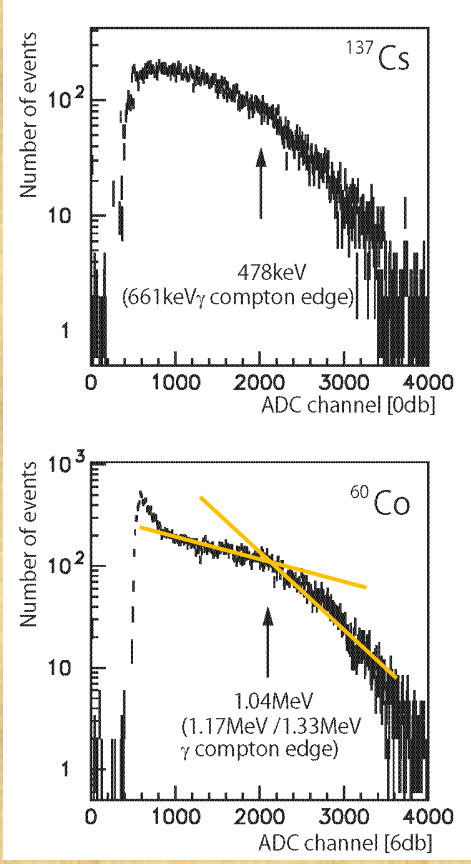
15250ch@0db

Quantum Yield :
15250/132800
=**11.5%(1.1%)**

Light Yield to BC505:
15250/208640
=7.3%

Response for γ–ray from radioactive source (2) : Quantum yield and light yield for AlQ_3

AlQ_3 in PhCN-POPOP



1040keV :
2025ch@6db

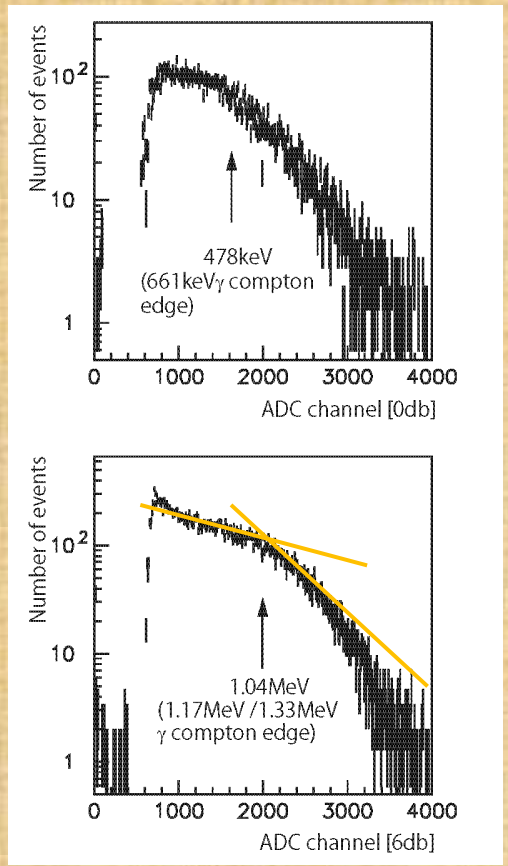
4050ch@0db
(QE : 0.126)

32143ch@0db

Quantum Yield :
32143/132800
=**24.2%(17.0%)**

478keV:
1885ch@0db
expected :
1861ch

AlQ_3 in PhCN-bis-MSB



1040keV :
1830ch@6db

3660ch@0db
(QE : 0.126)

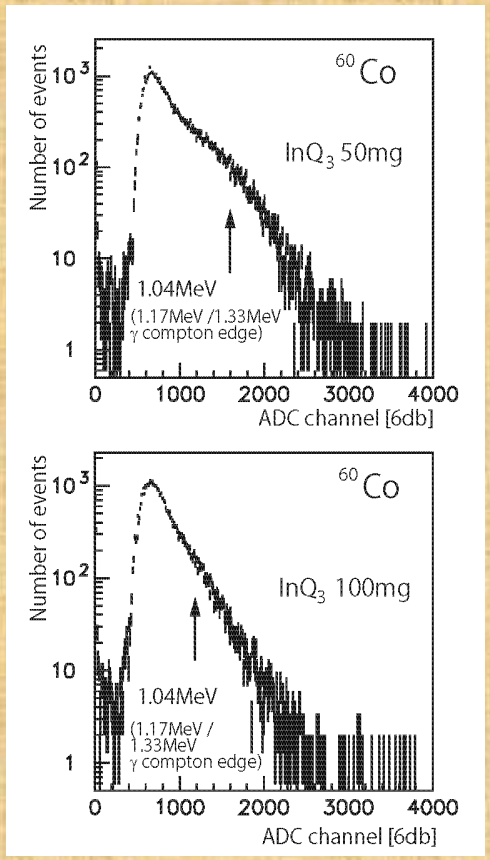
29048ch@0db

Quantum Yield :
29048/144320
=**20.1%(17.0%)**

478keV:
1625ch@0db
expected :
1682ch

Response for γ–ray from radioactive source (3) : effect for amount of complex

InQ_3 in PhCN-POPOP

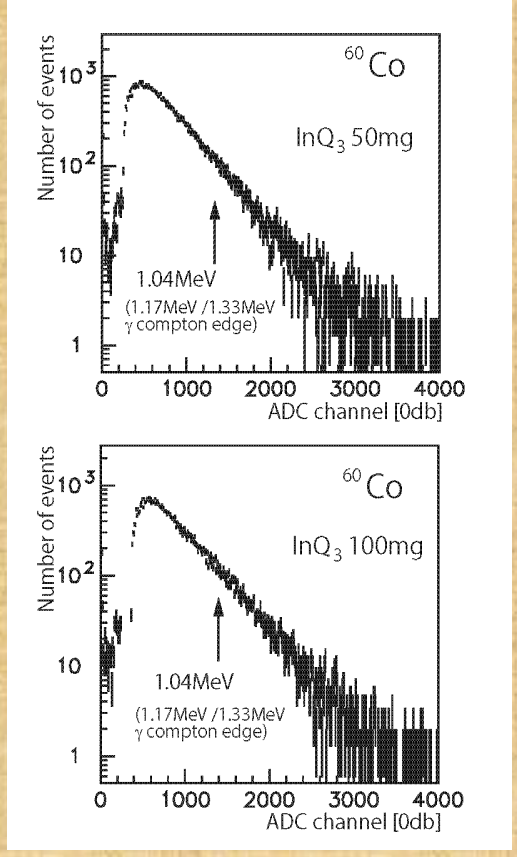


InQ_3 50mg
1040keV :
1530ch@6db

InQ_3 100mg
1040keV:
1080ch@6db

Light Yield :
△-29%

InQ_3 in PhCN + PPO 30mg



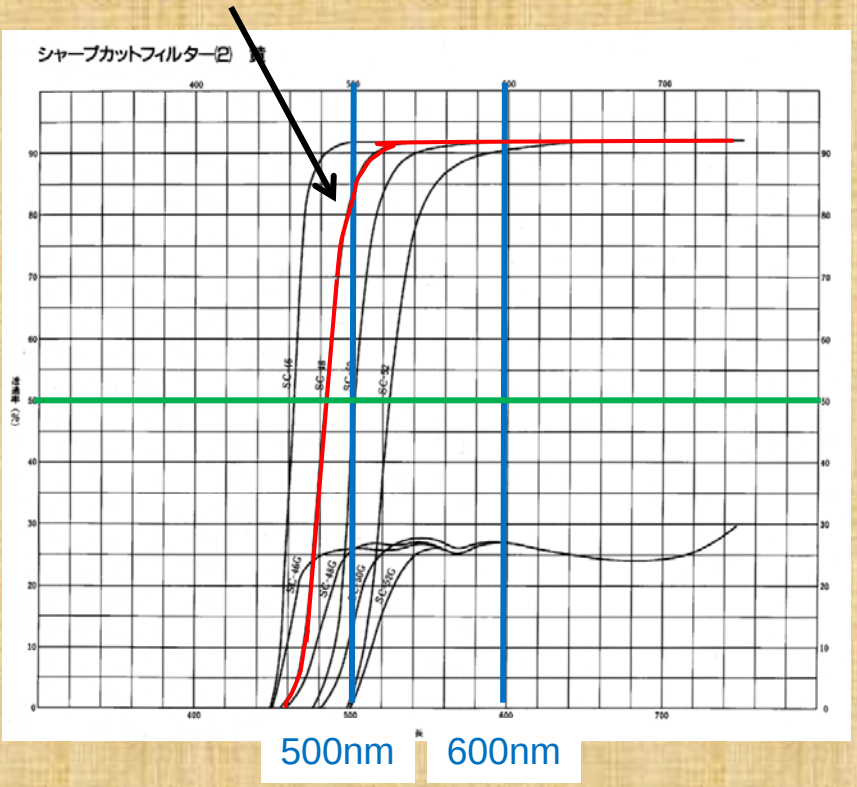
InQ_3 50mg
1040keV :
1030ch@0db

InQ_3 100mg
1040keV:
1030ch@0db

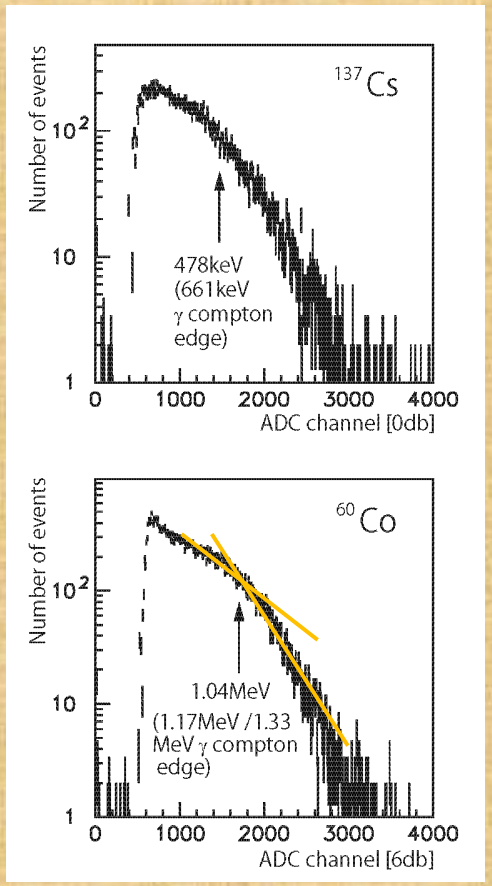
Light Yield :
△ ~0%

Response for γ–ray from radioactive source (4): PhCN scintillation light for λ > 530nm

FujiFilm SC-48 (λ > 530nm transparency : 92%)



PhCN-POPOP via SC-48



1040keV :
1525ch@6db

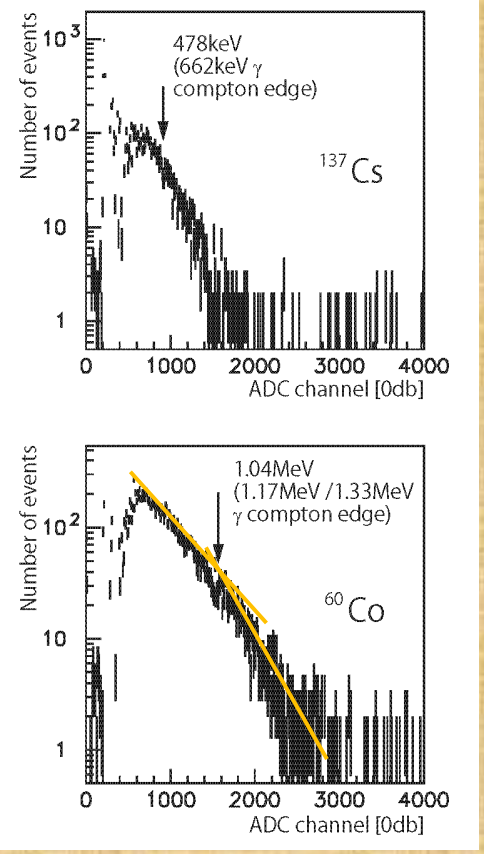
3050ch@0db
(QE : 0.10)

30500ch@0db

Light yield :
30500/132800
=23.0% remains

Response for γ–ray from radioactive source (5) : residual light in InQ_3 luminescence

InQ_3 50mg in PhCN-POPOP via SC-48



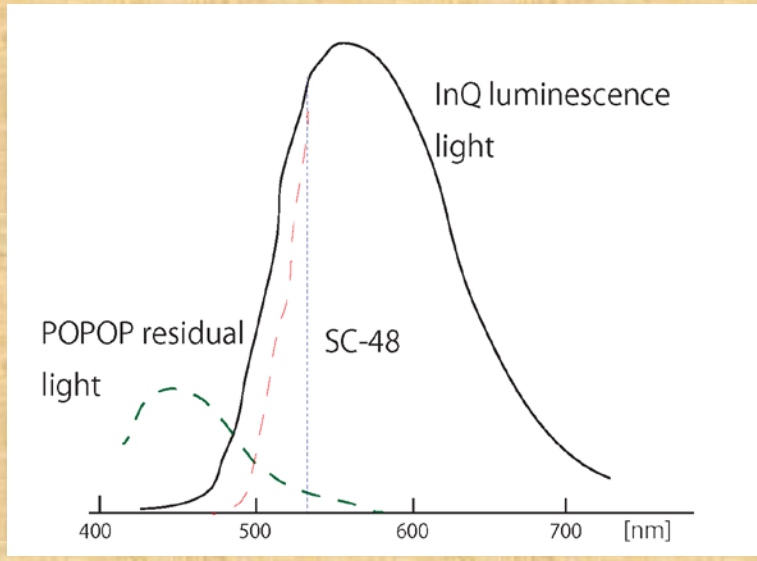
1040keV :
1280ch@0db
(QE : 0.093)

13763ch@0db

Light yield :
13763/20699
=66.5%

∴33.5% loss

residual lights in InQ_3 luminescence



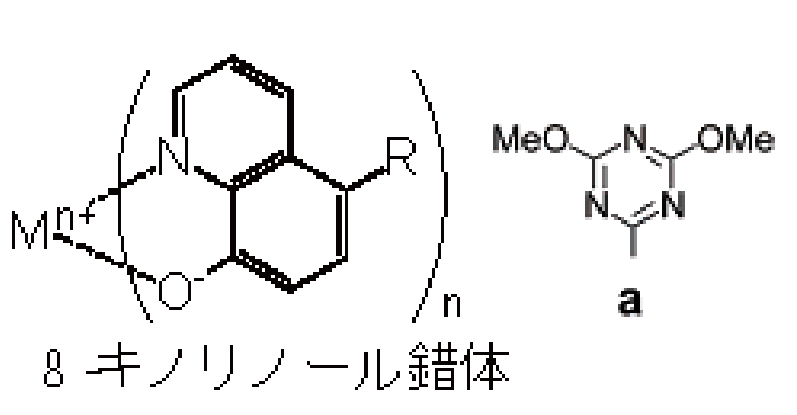
Most of the loss would be POPOP residual lights. It is consistent with the decrease of light yield for InQ_3 50mg → 100mg due to absorption of these light.

~10% of PhCN scintillation light remains

Results

- liquid scintillator containing tris(8-quinolinolate) metal complex (MQ_n) was made by PhCN as solvent and PPO100mg / POPOP(bis-MSB) 10mg
- InQ_3 and ZrQ_4 have photo luminescence for the γ radiation.
- Transparency : ~66cm @ 558nm (0.5% dissolution)
- Light yield relative to BC505 : 9.9% and 7.3%
- Quantum yield : 15.6% / 11.5% (PL: 5.0% / 1.1%)
- Next step: modify 8-quinolinolate ligand to add substituent groups in order to both increase QY and shorten wavelength or use another complex like β-diketone.

Tris(5-aryl-8-quinollinolate) metal complex



complex	A_{max} (ϵ [mol^{-1}·cm^{-1}])	λ_F [nm]	Φ_F[b]	τ_F [ns]	HOMO-LUMO gap [eV]
Alq_3	388 (7.0×10^3)	526	0.171	15.38	2.570
1a	390 (2.7×10^4)	490	0.533	29.50	3.255

- Expected light yield
  - Quantum Yield:

    InQ_3 0.05 →0.15~0.4

    ZrQ_4 0.01→0.03~0.37
  - emission wavelength:

    ~530nm for InQ_3

    QE 0.093→0.126

    ~524nm for ZrQ_4

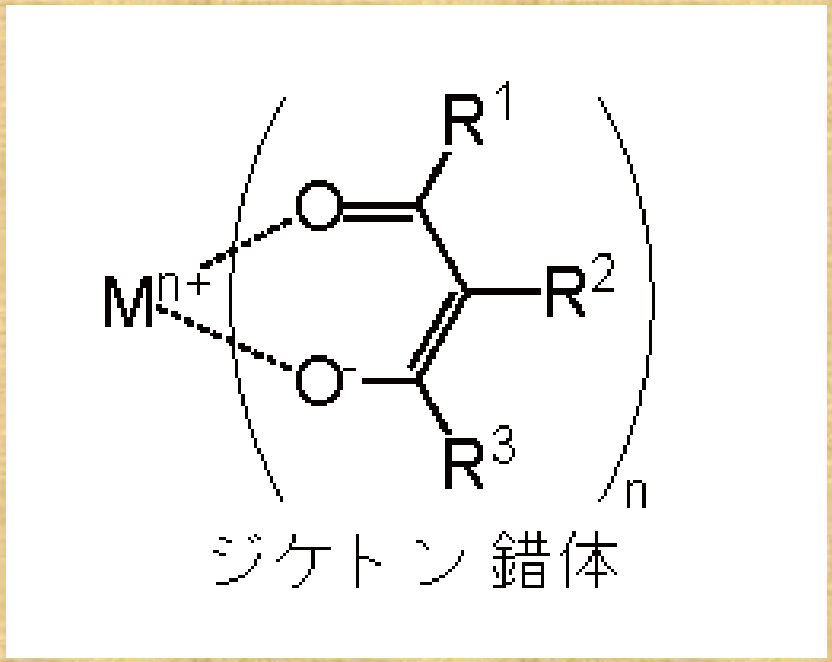
    QE 0.10→0.13

4~11 times to present light yield

β-diketone metal complex

- Good solubility to organic solvent

  10~20 wt.%
- Well established
- Easy synthesis
- Possible introduce substituent groups in order to move absorption wavelength



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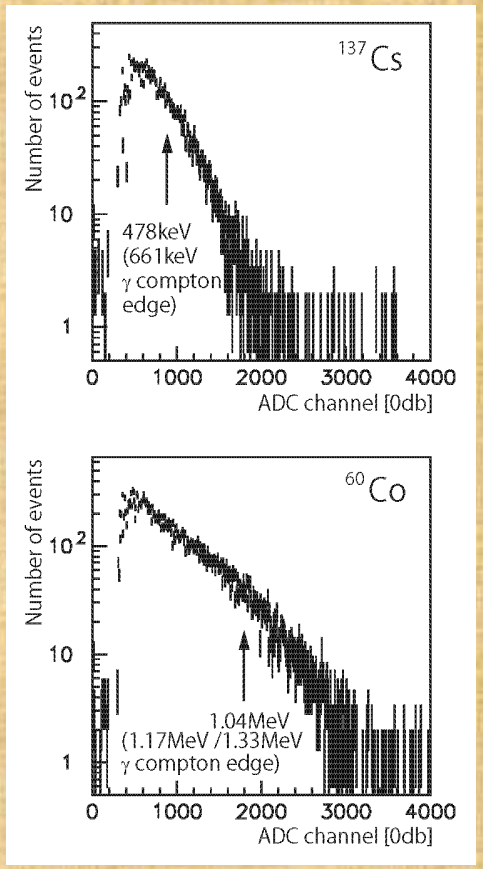
Backup

Response for γ–ray from radioactive source (6) : Quantum Yield and Light Yield

InQ_3 50mg in PhCN-bisMSB



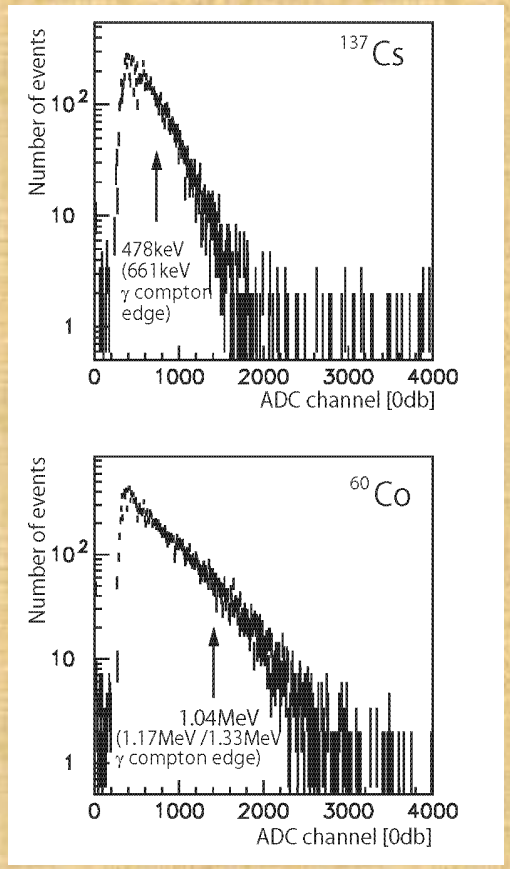
1040keV :
1630ch@0db
(QE : 0.093)

17527ch@0db

Quantum Yield :
17527/144320
=12.1%(5.0%)

Light Yield to BC505:
17527/208640
=8.4%

ZrQ_4 50mg in PhCN-bisMSB



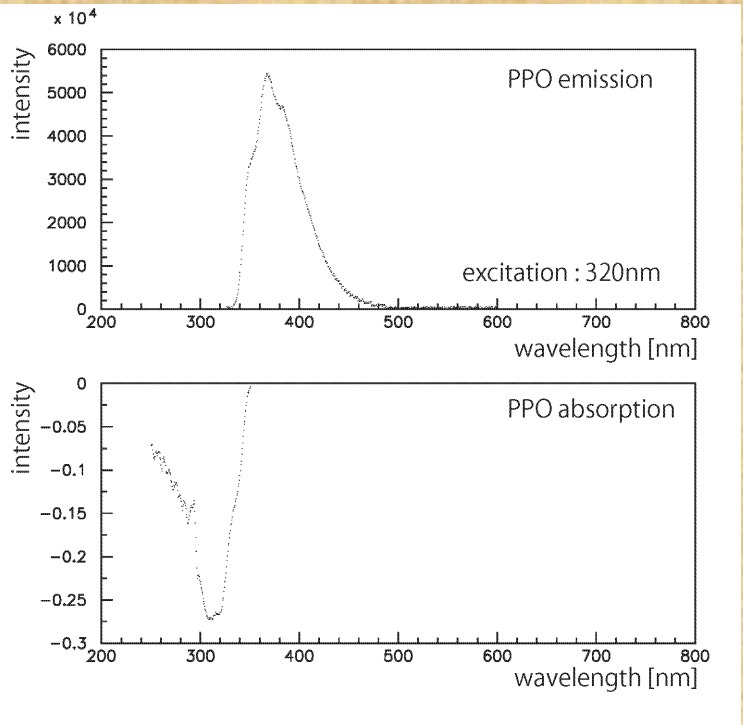
1040keV :
1230ch@0db
(QE : 0.10)

12300ch@0db

Quantum Yield :
12300/144320
=8.5%(1.1%)

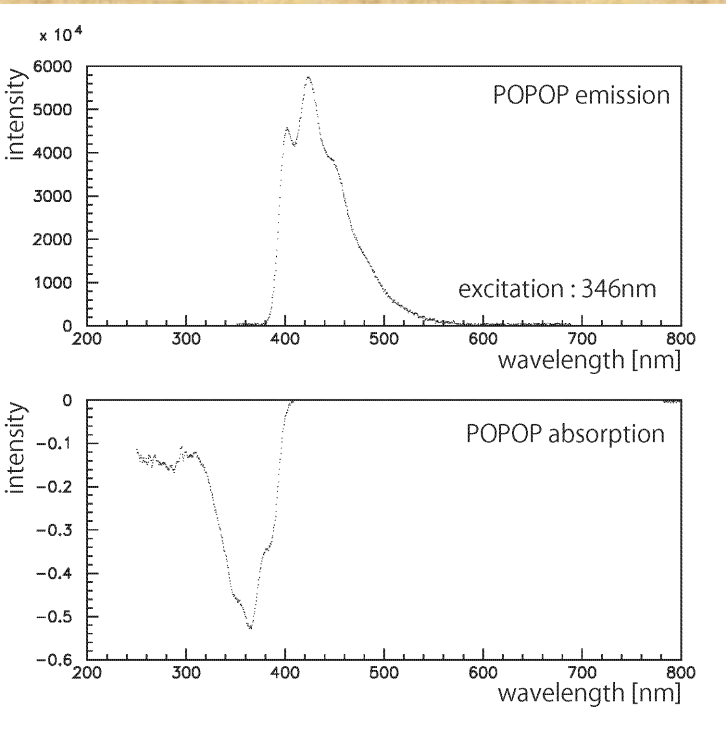
Light Yield to BC505:
12300/208640
=5.9%

Photo Luminescence and absorption of PPO



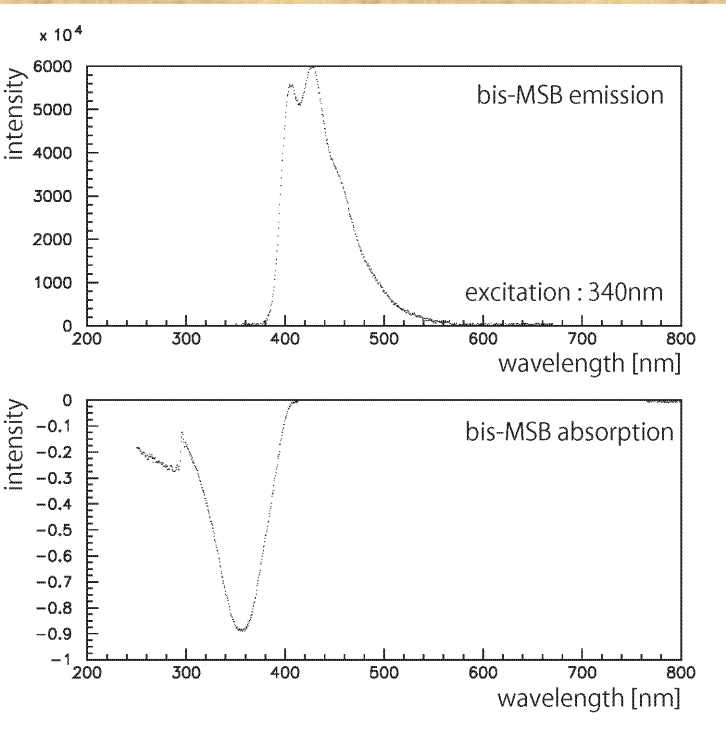
- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance devie : HITACHI U-3000
  - Solvent : Benzonitrile (PhCN)
  - Concentration : 1.0×10^{-5} mol/L
  - 2,5-Diphenyloxazole
  - Molecular mass : 221.26
  - Max. emission wavelength : 368.0nm
  - Max. absorption wavelength : 309.7nm

Photo Luminescence and absorption of POPOP



- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance devie : 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 devie : 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

Synthesis of InQ_3