Optical characterization of the liquid scintillator at low temperatures

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Outline

- main skills and activities of the group
- devices for low temperature measurements
- previous results and a few literature data
- pure LAB
- LAB + PPO + BisMSB
- old recipe for TAO
- latest TAO recipe and possible new cocktails
- conclusions

Main skills and activities of the group

➢ **Our group is mainly involved in the activities related to the liquid scintillator**

a) **Set-up, characterization, stability check of the liquid scintillator and study of the possible interactions with the materials employed for the detector, carried out in the UV and visible regions**

b) **Possibility to improve the optical characteristic of the liquid scintillator**

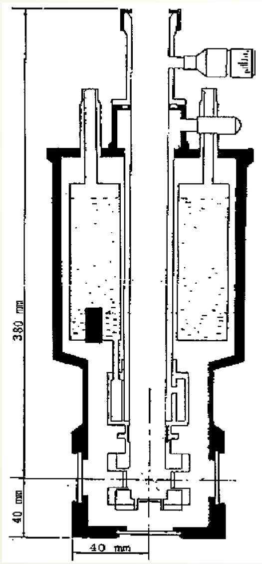
c) **Optical measurements in cryogenic conditions**

Instrumentation

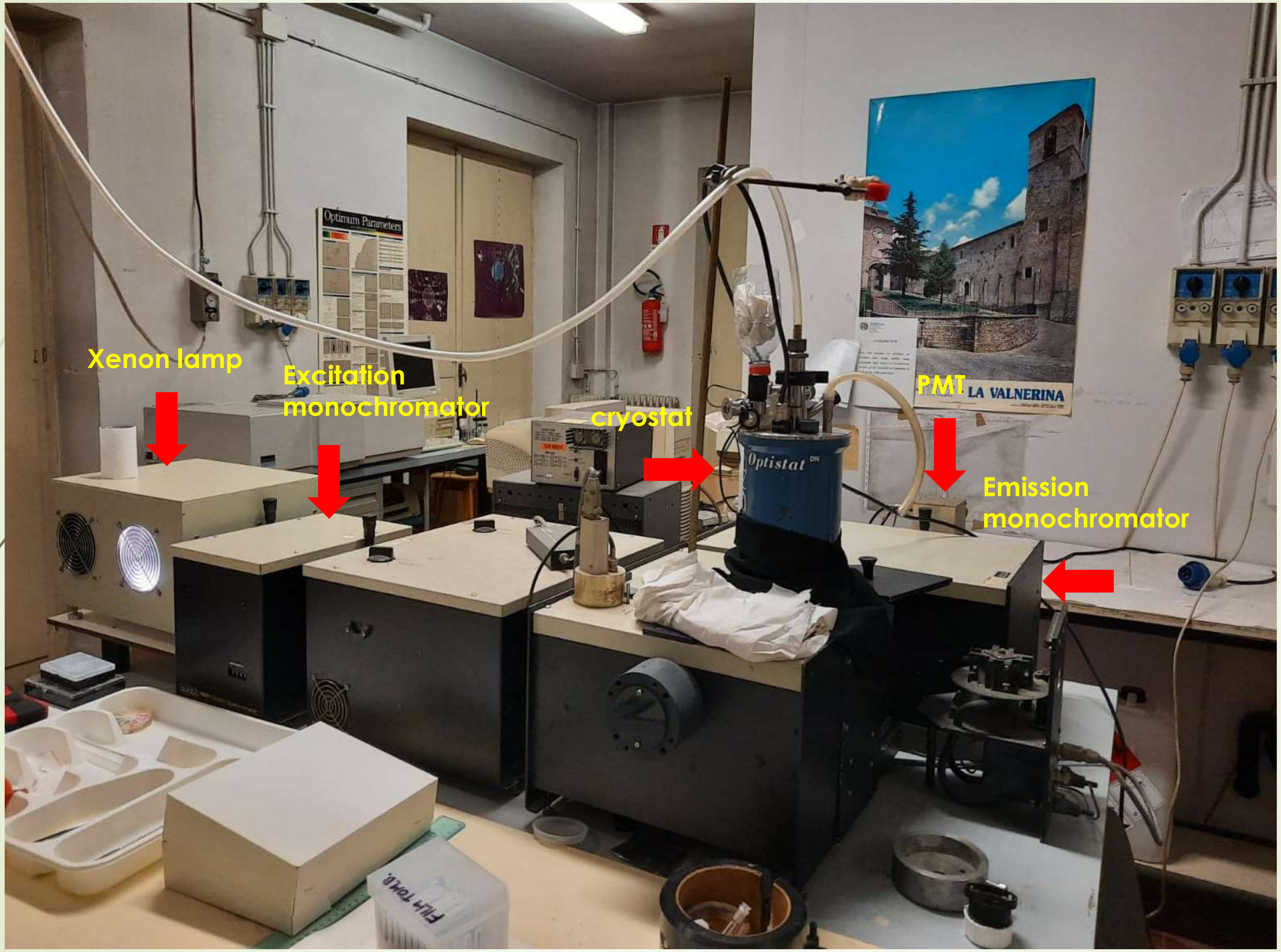
Absorption measurements

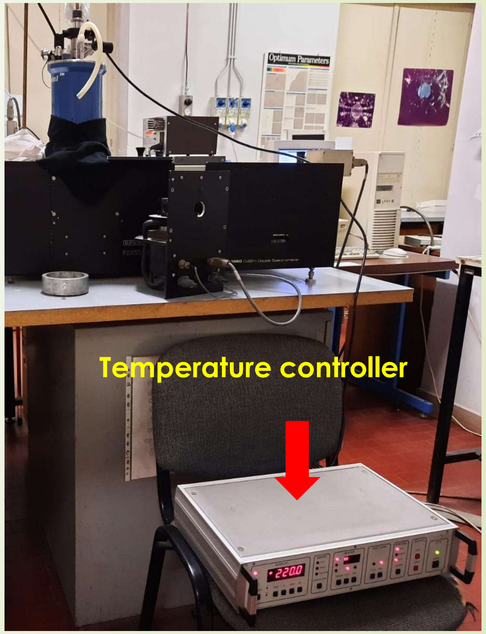
- **Absorption spectra (optical path from 1 to 10 cm)**
- **Attenuation length (optical path 10 cm)**
- **Measurements at room temperature and under cryogenic conditions**

cryostat

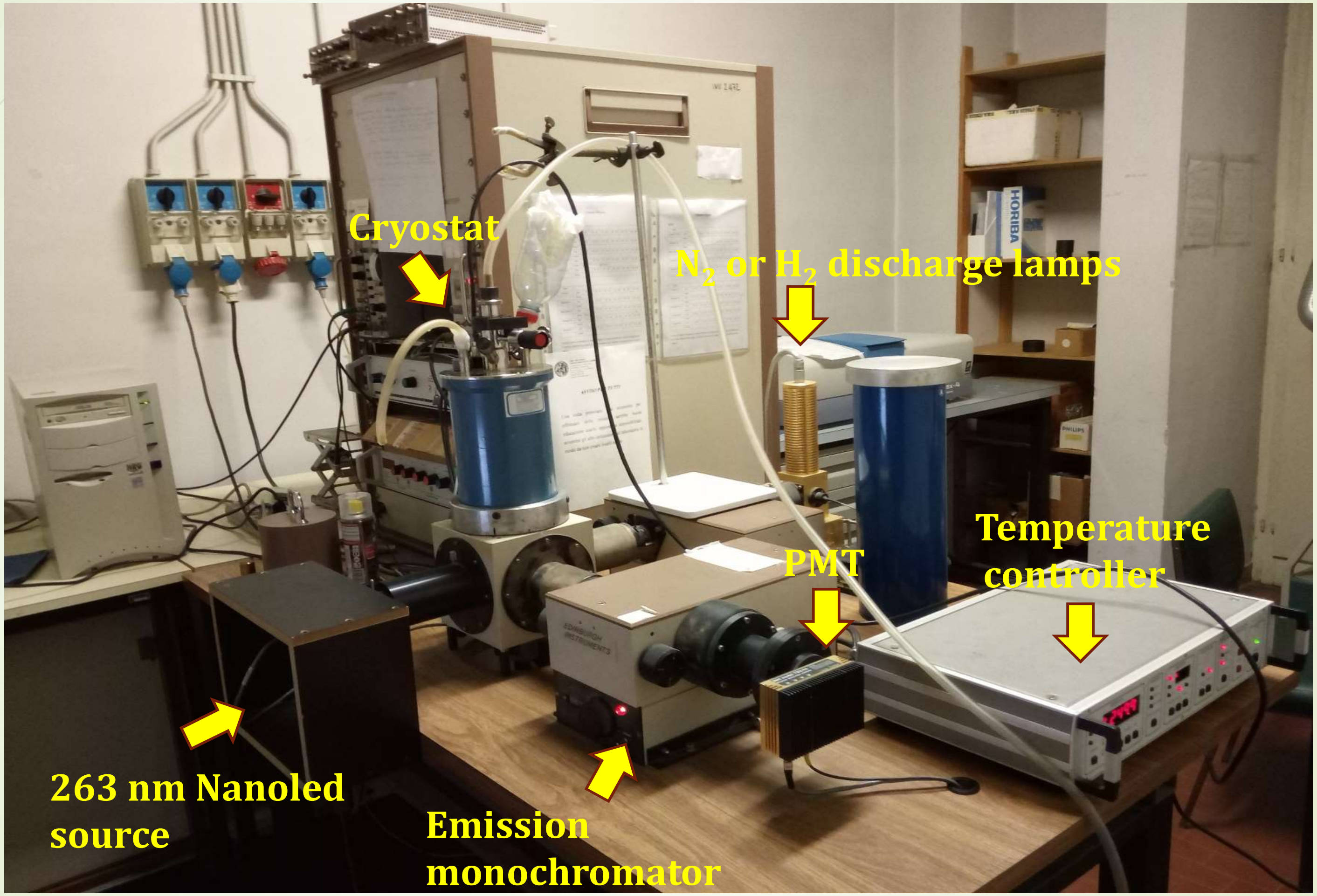


Emission measurements





Fluorescence decay lifetime measurements



Previous results and a few literature data

ROUND-TABLE ON
'IMPROVEMENT OF LIQUID SCINTILLATOR RESPONSE'

HOWARD H. SELIGER
National Bureau of Standards, Washington, D.C.

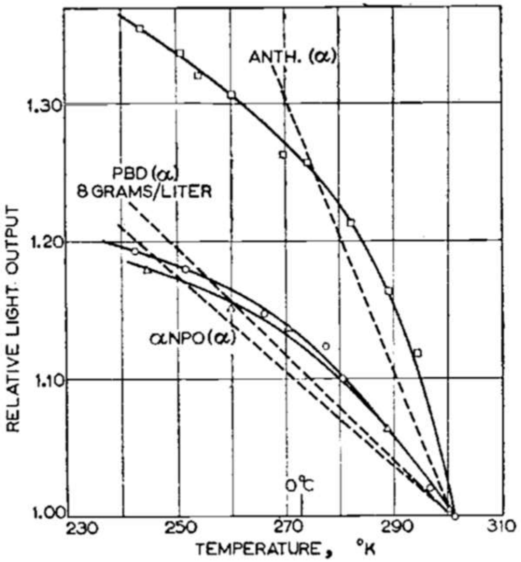
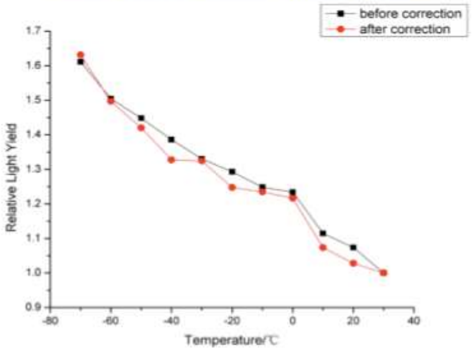


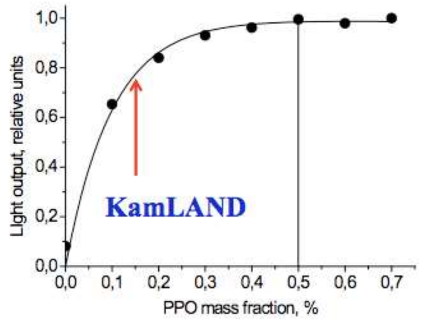
Fig. 7. Relative light outputs vs. temperature for gas-free liquid scintillators. The dashed curves represent data for alpha excitation. The solid curves represent the data for gamma excitation.
□, Anthracene 1 g/l.; ⊙, PBD 10 g/l. + POPOP 2 g/l.; △, α-NPO-3, 2 g/l.

More Photons-- LS

- **Longer attenuation length**
  - ⇨ Improve raw materials (using Dodecane instead of MO for LAB production)
  - ⇨ Improve the production process
  - ⇨ Purification
- **Higher light yield**
  - ⇨ Lower temperature
  - ⇨ fluor concentration optimization

Linear Alky Benzene	Atte. Length @ 430 nm
RAW	14.2 m
Vacuum distillation	19.5 m
SiO_2 coloum	18.6 m
Al_2O_3 coloum	22.3 m





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Int. J. Radiat. Appl. Instrum. Part A

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The Effect of Temperature on Fluorescence for Liquid Scintillators and Their Solvents

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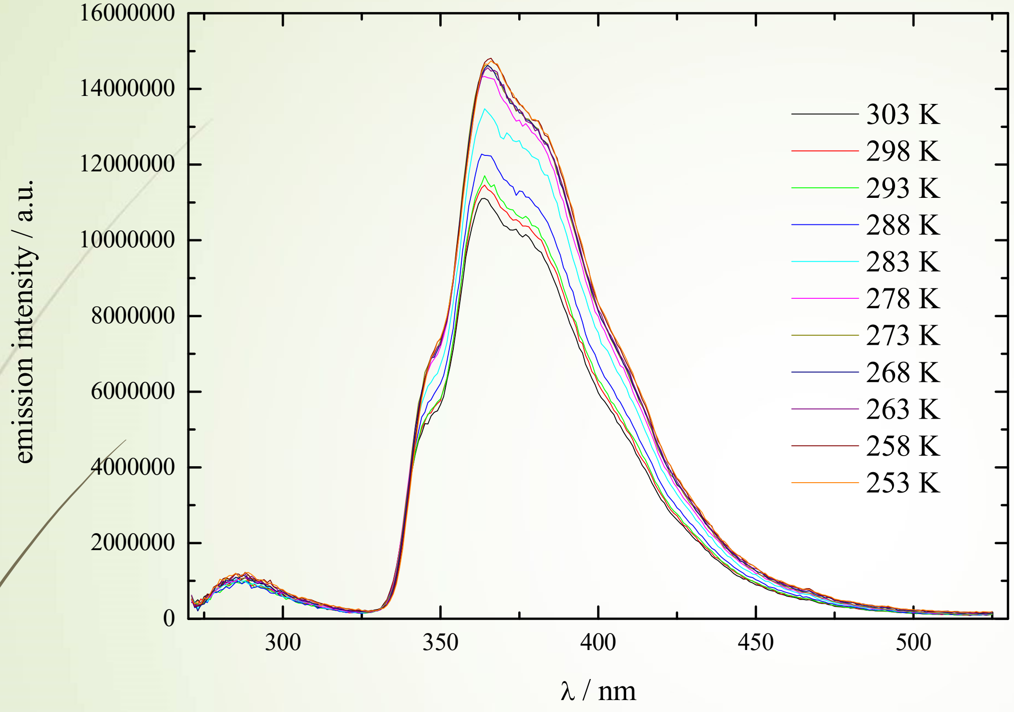
(*Received* 16 *December* 1985; *in revised form* 29 *August* 1986)

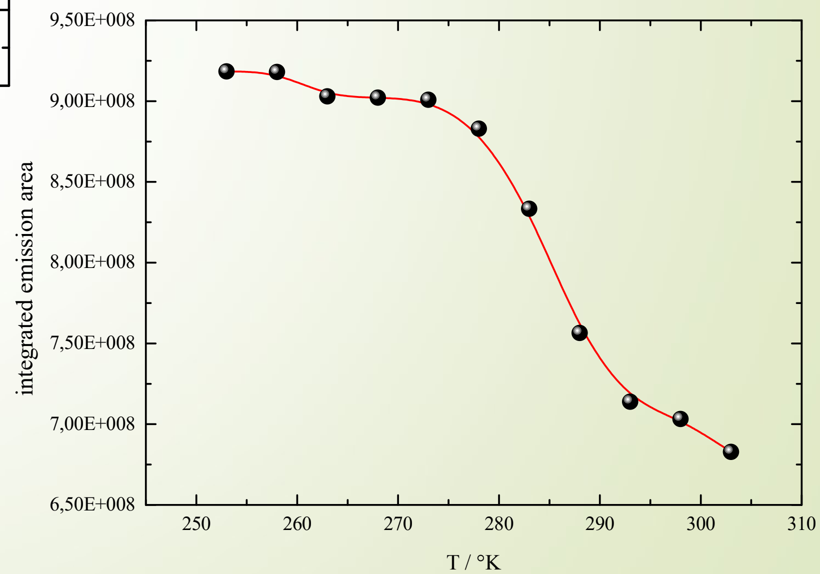
The pulse-height distributions for ^{241}Am and ^{131m}Xe in PPO solutions of the aromatic hydrocarbons, i.e. benzene, *o*-xylene, *m*-xylene, ethylbenzene and cumene are found to shift toward higher pulse-heights with decreasing temperature. Under u.v. excitation, the fluorescence intensity of these pure aromatic hydrocarbons increases markedly and the fluorescence maximum is found to shift to longer wavelengths with decreasing temperature.

In conjunction with observations of the pulse-height shift in liquid scintillators and the fluorescence emission from the pure solvents, the pulse-height shifts observed are interpreted in terms of excimer formation in the aromatic hydrocarbons.

Measurements for the Borexino experiment



- **Emission spectra recorded on the Borexino scintillator (pseudocumene + 1.5 g/l PPO)**



- **Temperature range investigated: 303-253 K**

Optical measurements on pure LAB

- **Importance related to the possible use of SiPM in the near detector at low temperature**
- **several possible issues:**

- is LAB still fluid at low T? Melting point of benzene is about 5°C, whilst that of toluene, the simplest alchil-benzene is -95°C, what about other LABs? For dodecyl-benzene mp = -7°C

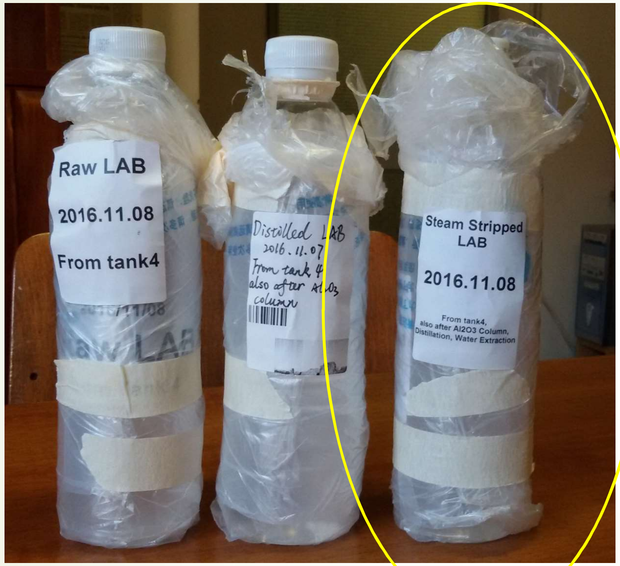
- LAB is usually a mixture of isomers, ranging from C9 to C13, probably a melting interval, rather than a melting point, is expected

- if LAB undergoes distillation, the composition may change, increasing the percentage of lighter isomers in the mixture, therefore mp may vary as well

- when solutes are added (PPO, bisMSB, etc.) melting point may change

- solubility usually decreases upon decreasing temperature: possible PPO precipitation?

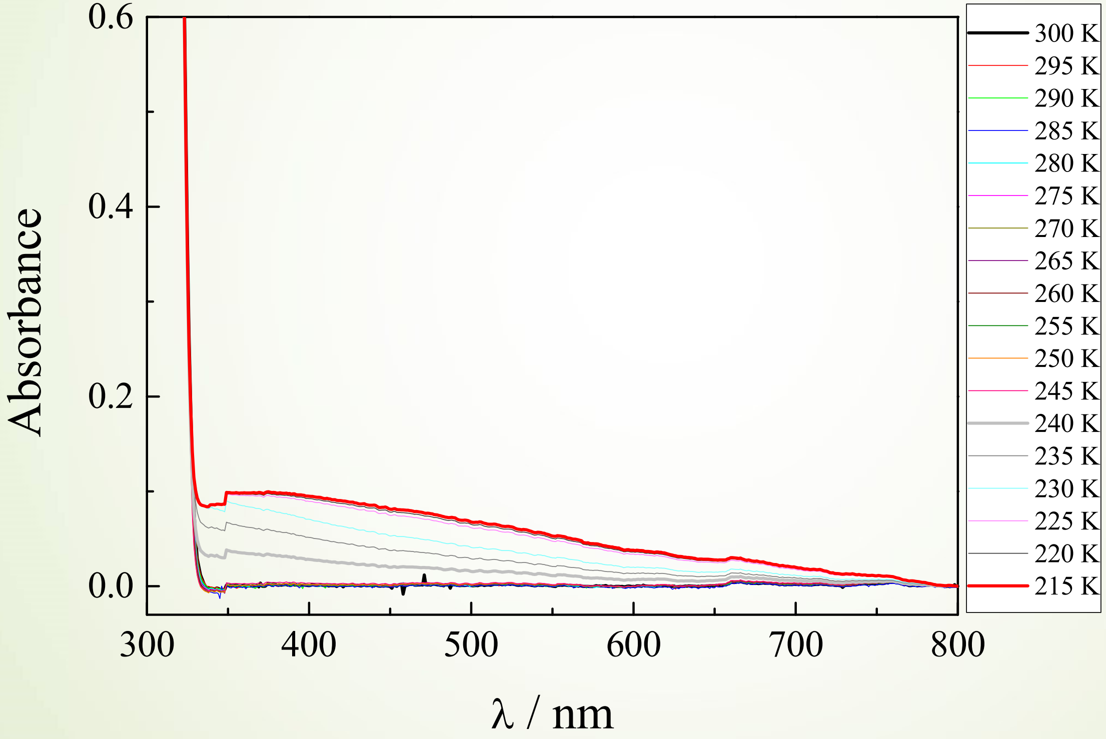
➢ a **LAB sample from the tank 4 of the DayaBay plant**, which had undergone Al_2O_3 column treatment, distillation, water extraction and steam stripping, was used for this test



➢ measurements were carried out on a quartz cell, CCl_4 as reference

➢ the sample was purged by nitrogen bubbling for 15 minutes, and then sealed with Teflon and Parafilm, before the test

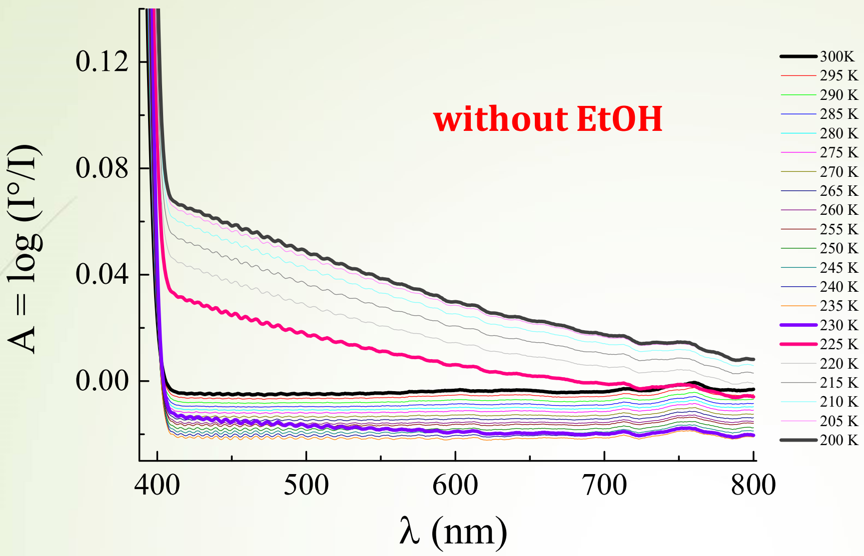
- spectra were recorded in the temperature interval from 300 K down to 215 K
- at 240 K the spectral profile changed and a light opalescence appeared. This “critical” temperature was checked and confirmed the following day using a fresh sample and going directly down to 250 K

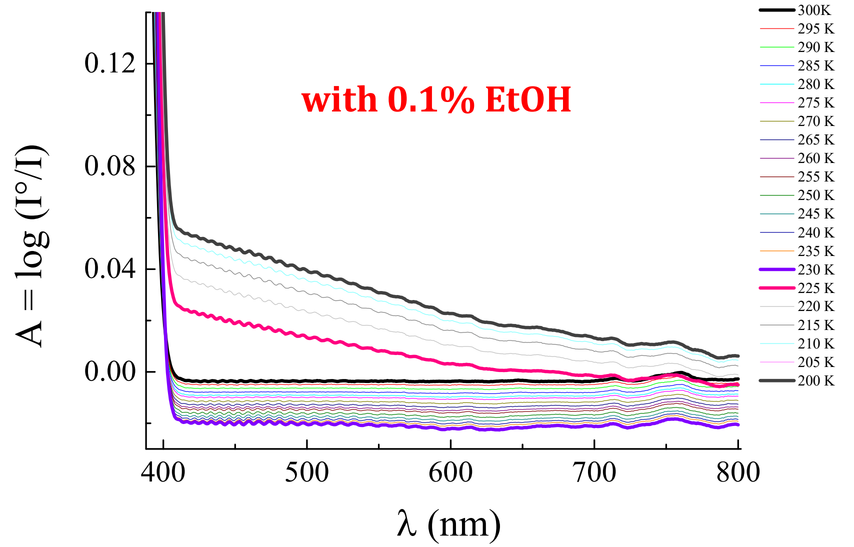


- at the lowest temperature explored (215 K) the sample was still fluid, even though it exhibited a higher viscosity, as expected
- the sample was left in the cryostat overnight, without any heating source, in order to reach the lowest temperature possible, that is the nitrogen boiling point (77 K)
- at this temperature the sample was solid and had completely lost its transparency
- residual water in the sample due to water extraction procedure?

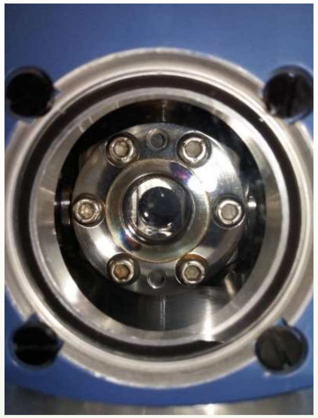
Optical measurements on LAB + PPO + BisMSB

- **Measurement of the absorption properties of LAB (after treatment with Al_2O_3 column, distillation, water extraction and steam stripping) with PPO (3 g/l) and bisMSB (1-2 mg/l) were carried out under the same experimental conditions**
- **Taking into account previous JUNO-TAO tests, a sample of the same LS, modified with 0.1% dried ethanol was also prepared and investigated**
- **The samples were purged by nitrogen bubbling for 15 minutes, and then sealed with Teflon and Parafilm tapes, before the test**

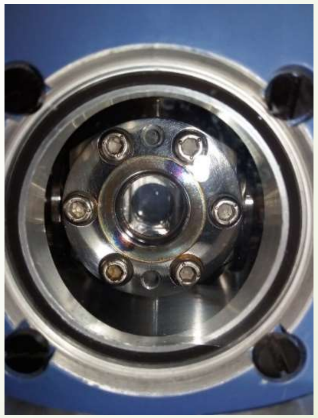




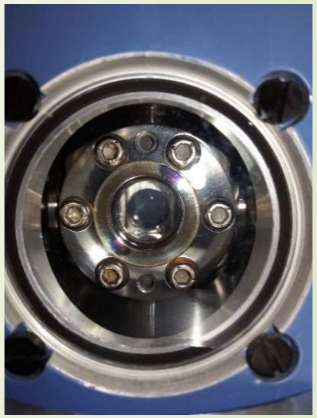
300 K



230 K



225 K



Pictures taken from the front window of the cryostat

- The addition of fluors to pure LAB improves the optical transmittance of the LS at low temperatures

- Addition of 0.1% ethanol also seems to positively affect the transparency at low T, even though the effect is barely perceptible

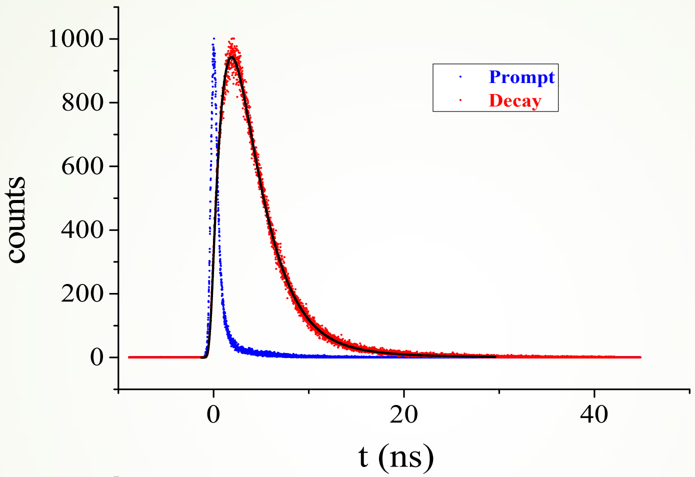
- Removing the cell from the cryostat, at the end (T = 200 K) LS is highly viscous, but still fluid

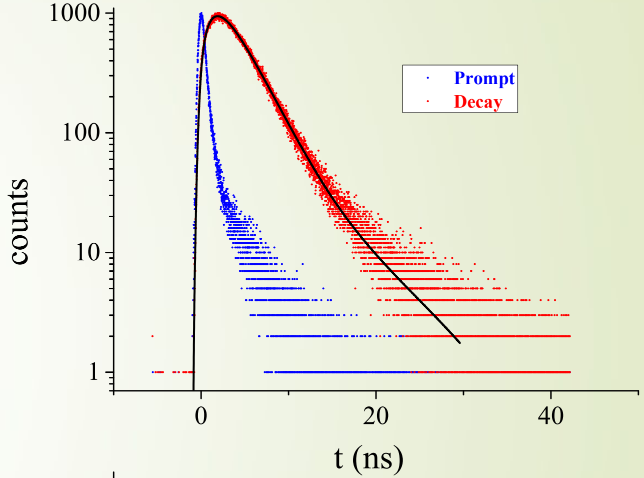
- Some opalescence is clearly visible

Low T lifetime measurements on LS (LAB +0.1% dried EtOH + PPO + bisMSB)

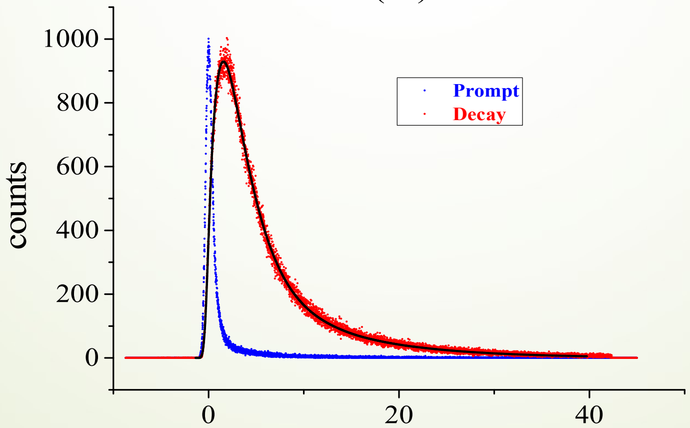
- **sample degassed by N_2 for 15'**
- **nanoled source @263 nm for UV excitation**
- **emission monochromator set @ 420 nm for emission**

300 K

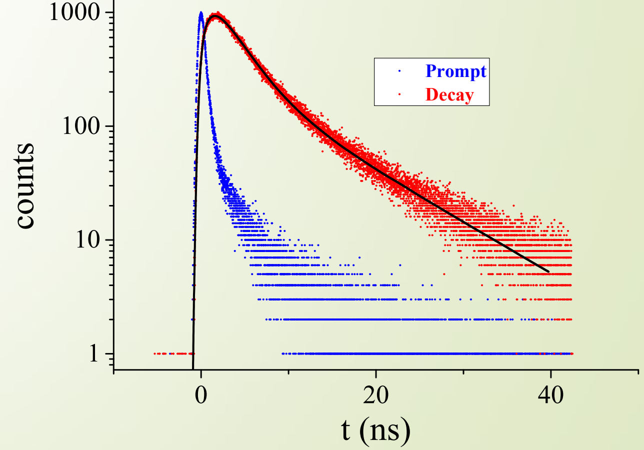




200 K



t (ns)



Summary of lifetime results

	r.t (ns)	τ_1 (ns)	τ_2 (ns)
300 K	1.84 ± 0.06	1.85 ± 0.06	7.0 ± 0.2
295 K	1.83 ± 0.05	1.84 ± 0.06	7.2 ± 0.2
290 K	1.85 ± 0.06	1.85 ± 0.06	7.6 ± 0.2
285 K	1.86 ± 0.06	1.86 ± 0.06	7.9 ± 0.2
280 K	1.87 ± 0.06	1.89 ± 0.06	8.1 ± 0.2
275 K	1.83 ± 0.05	1.86 ± 0.06	7.8 ± 0.2
270 K	1.86 ± 0.06	1.86 ± 0.06	7.9 ± 0.2
265 K	1.51 ± 0.05	2.26 ± 0.07	8.3 ± 0.2
260 K	1.82 ± 0.05	1.84 ± 0.06	7.7 ± 0.2
255 K	1.37 ± 0.04	2.34 ± 0.07	8.3 ± 0.2
250 K	1.31 ± 0.04	2.40 ± 0.07	8.5 ± 0.3
245 K	1.26 ± 0.04	2.53 ± 0.08	9.0 ± 0.3
240 K	1.21 ± 0.04	2.46 ± 0.07	8.7 ± 0.3
235 K	1.09 ± 0.03	2.64 ± 0.08	9.1 ± 0.3
230 K	1.15 ± 0.03	2.48 ± 0.07	9.1 ± 0.3
225 K	1.08 ± 0.03	2.45 ± 0.07	9.1 ± 0.3
220 K	1.10 ± 0.03	2.41 ± 0.07	9.2 ± 0.3
215 K	1.12 ± 0.03	2.32 ± 0.07	9.4 ± 0.3
210 K	1.10 ± 0.03	2.32 ± 0.07	9.5 ± 0.3
205 K	1.10 ± 0.03	2.28 ± 0.07	9.5 ± 0.3
200 K	1.05 ± 0.03	2.35 ± 0.07	9.8 ± 0.3

- **the rise time decreases upon decreasing T**
- **both the short- and the long-lived components of the decay slightly increase upon decreasing T**

Measurements on the old recipe for TAO

- The sample was prepared following the old TAO recipe: **LAB + 2 g/L PPO + 1 mg/L bisMSB + ~ 0.05% ethanol**.

- A LAB sample from tank 4 of the DayaBay plant, which had undergone Al_2O_3 column treatment, distillation, water extraction and steam stripping, was used.

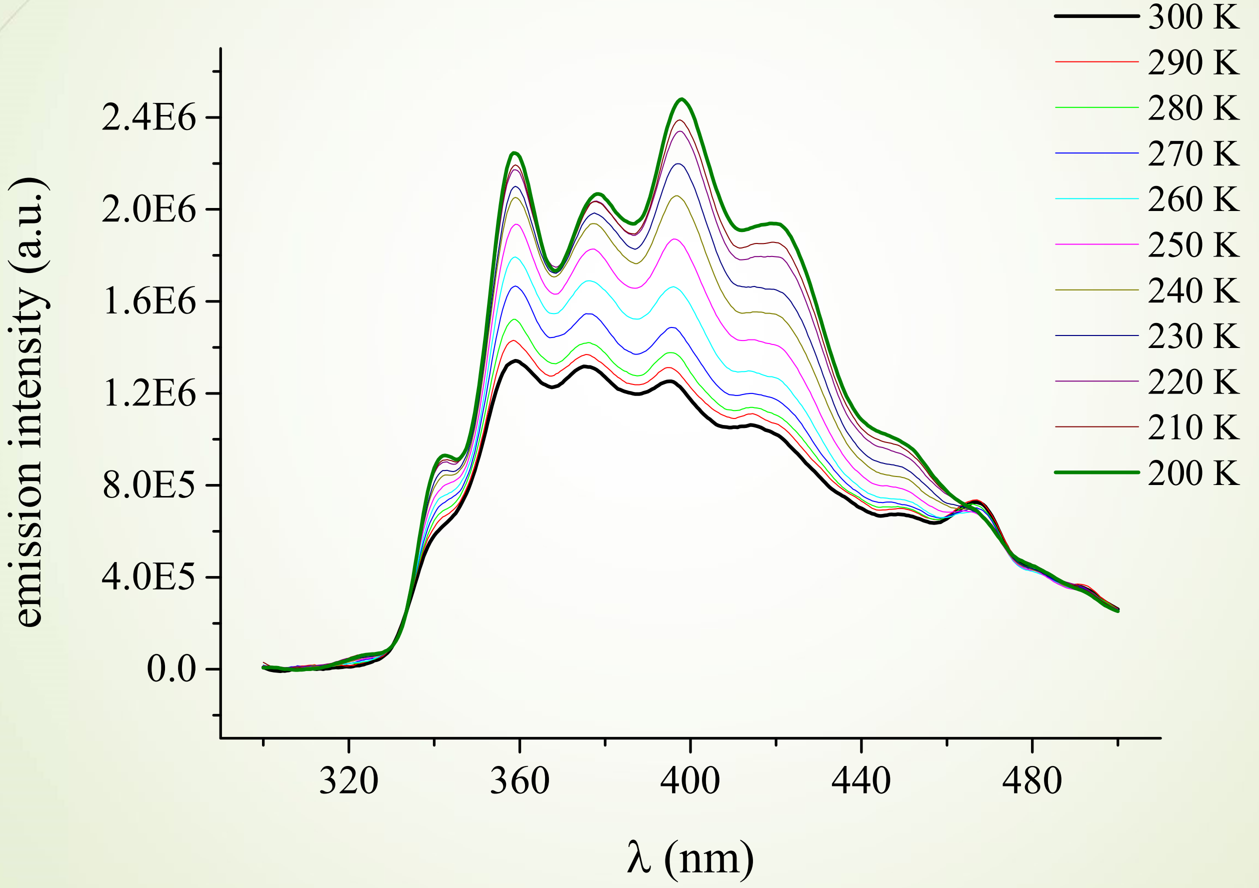
- Two cuvettes, one containing the solution of LAB + PPO + bisMSB and the other filled with anhydrous ethanol were sealed and separately purged for 20 minutes by nitrogen flux using silicone rubber caps and needles for gas inlet and outlet.

- 1.3 microliters of degassed ethanol were withdrawn by means of a syringe and injected into 2.5 ml of the scintillator solution, to obtained the desired composition.

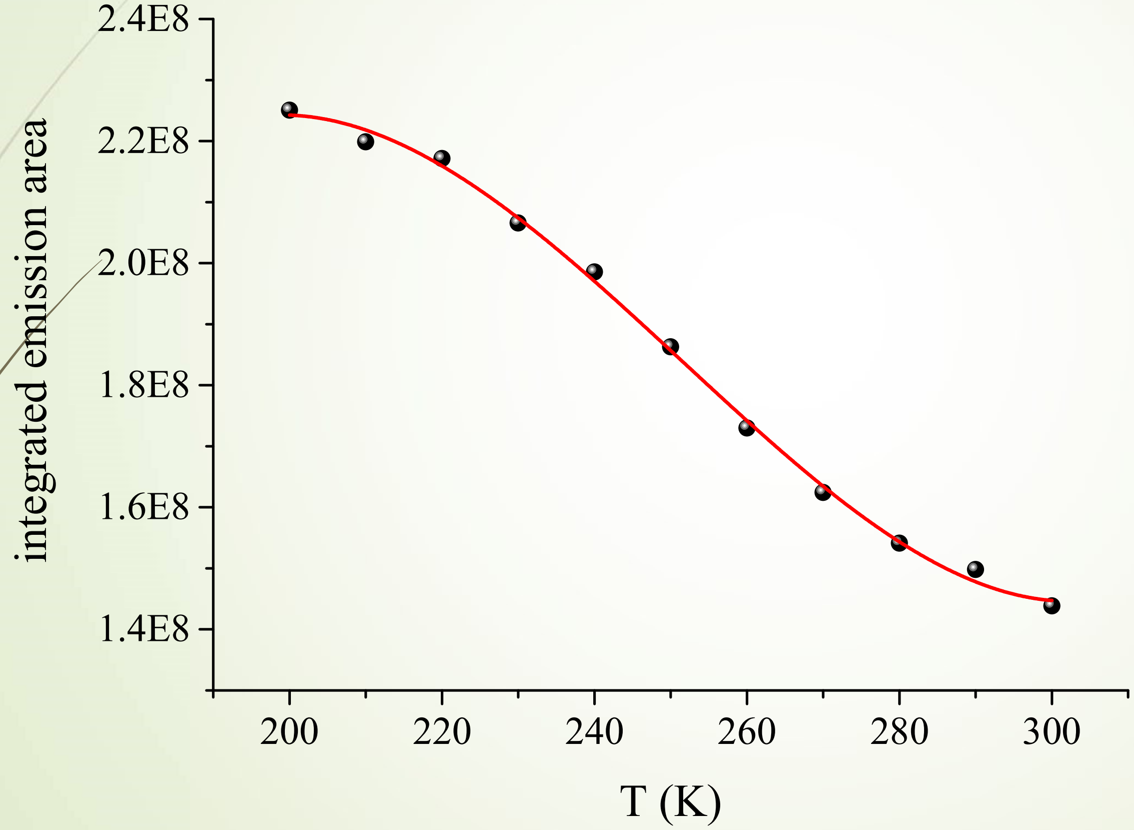
- This procedure was followed in order to avoid selective ethanol evaporation that would occur upon fluxing the solution containing all the components, due to the large difference of LAB and ethanol boiling points.

Emission spectra vs temperature (300 K – 200 K)

- **The emission spectra, recorded upon excitation of the sample at λ = 265 nm, were collected in the 300 K – 200 K temperature range, at intervals of 10 degrees.**



Relative light yields vs temperature (300 K – 200 K)



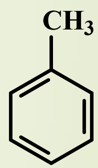
- **The integrated emission areas regularly increased with decreasing temperature, in the 300 K – 200 K range.**
- **More than 40% increase in the emission yield was observed going from room temperature (about 290 K) down to 220 K.**
- **The effect might be somehow overestimated due to volume contraction at low T.**
- **This issue is probably less important working in a front-face geometry, compared to a classical 90-degree mode.**

Latest TAO recipe and possible new cocktails

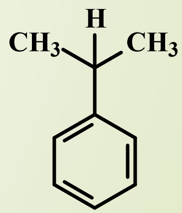
- **The materials used for sample preparation were kindly provided by Dr. Hans Steiger**
- **The first sample was based on a Gd-loaded LAB prepared at IHEP, upon addition of 3 g/l PPO, 2 mg/l BisMSB and 0.5% DPnB as co-solvent (TAO LS recipe)**

Two alternative compositions were also tested:

- **Helm LAB loaded with 3 g/l PPO, 2 mg/l BisMSB and 0.5% toluene as co-solvent**

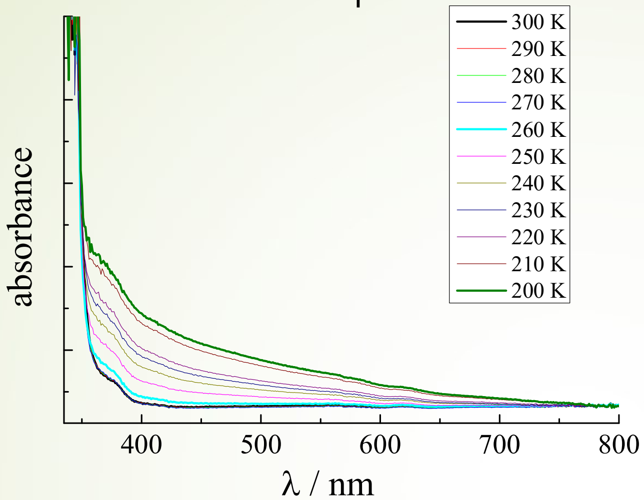


- **Helm LAB loaded with 3 g/l PPO, 2 mg/l BisMSB and 0.5% cumene as co-solvent**

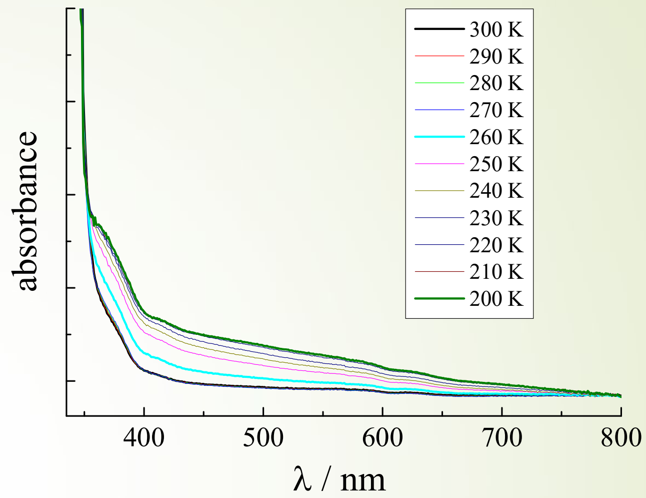


Absorption spectra as a function of temperature (300 K – 200 K)

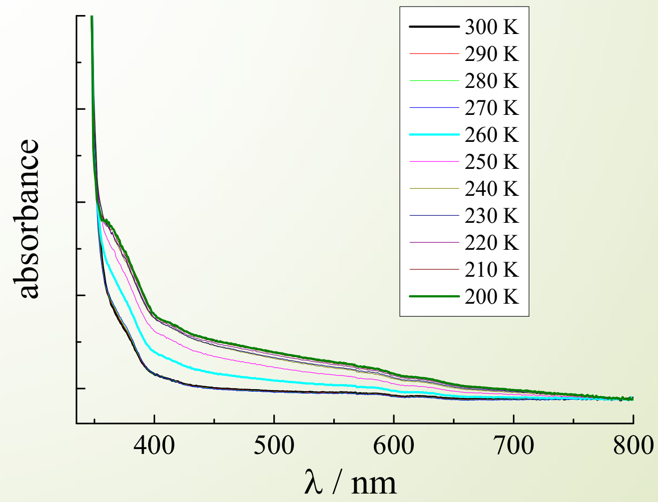
TAO recipe



Toluene as co-solvent



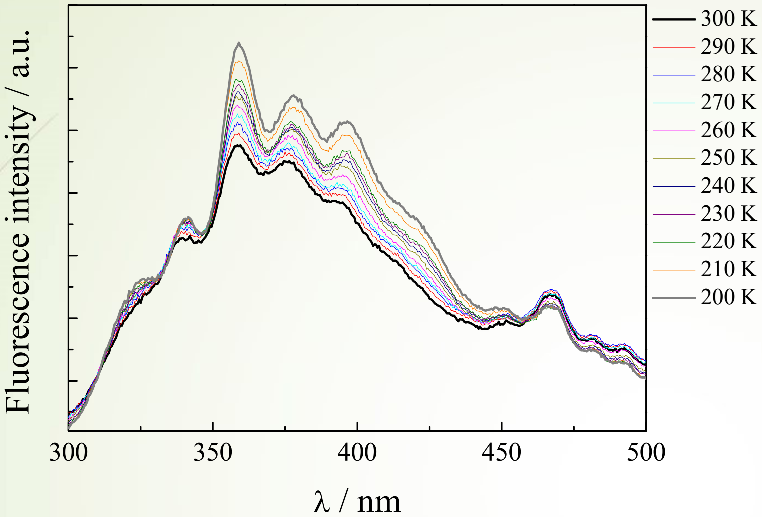
Cumene as co-solvent



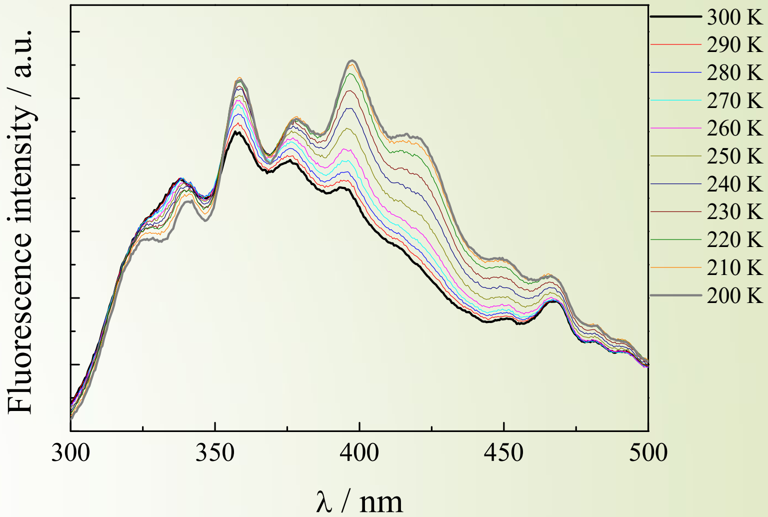
- **All samples exhibit the presence of scattered light at 260 K, that increases further upon decreasing T**
- **The reason is probably the presence of water traces rather than precipitation of solutes**
- **Old tests performed with higher content of PPO and BisMSB and ethanol as co-solvent had shown the onset of scattered light at lower T (225-230 K)**

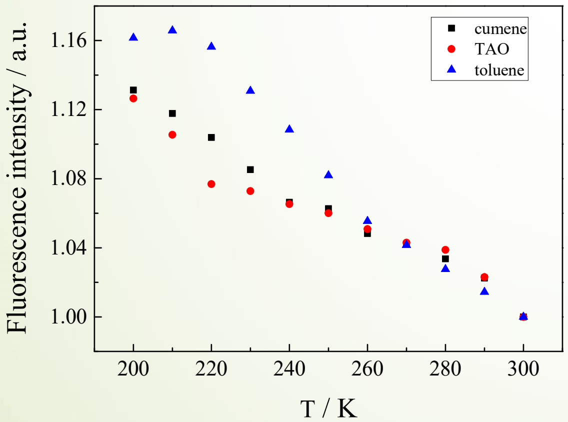
Emission spectra as a function of temperature (300 K – 200 K)

TAO recipe

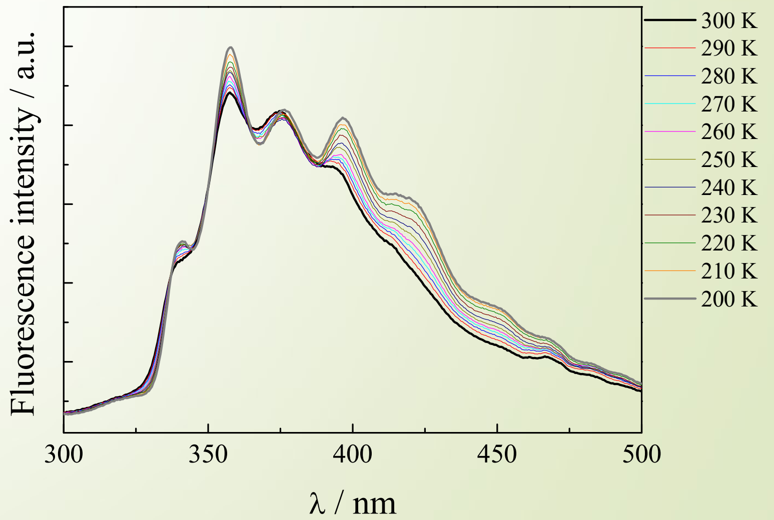


Toluene as co-solvent





Cumene as co-solvent



- **The LS containing 0.5% toluene exhibits a higher relative increase of the fluorescence light upon decreasing temperature**

Conclusions

- **Optical measurements on scintillating mixtures of various compositions, as a function of temperature (300 K - 200 K), from pure LAB to the latest TAO recipe, containing 0.5% DPnB as a co-solvent, to end up with two possible alternative cocktails, where DPnB has been replaced with either toluene or cumene.**

- **The last tests show the presence of scattered light from 260 K, instead of 235-240 K, found from previous analyses, that makes worse with decreasing temperature.**

- **Traces of residual water in the samples (?), rather than precipitation of the solutes dissolved in LAB.**

- **Increase of emission light yield upon decreasing temperature, as expected.**

- **As for the alternative scintillating cocktails, the solution containing 0.5% toluene exhibits a larger percentage increase of the light yield with decreasing temperature.**

- **Absorption, emission and lifetime measurements on other possible solutions containing other alternative co-solvents might be of some interest.**