Toward a standardization of scintillating performances of nanoscintillators

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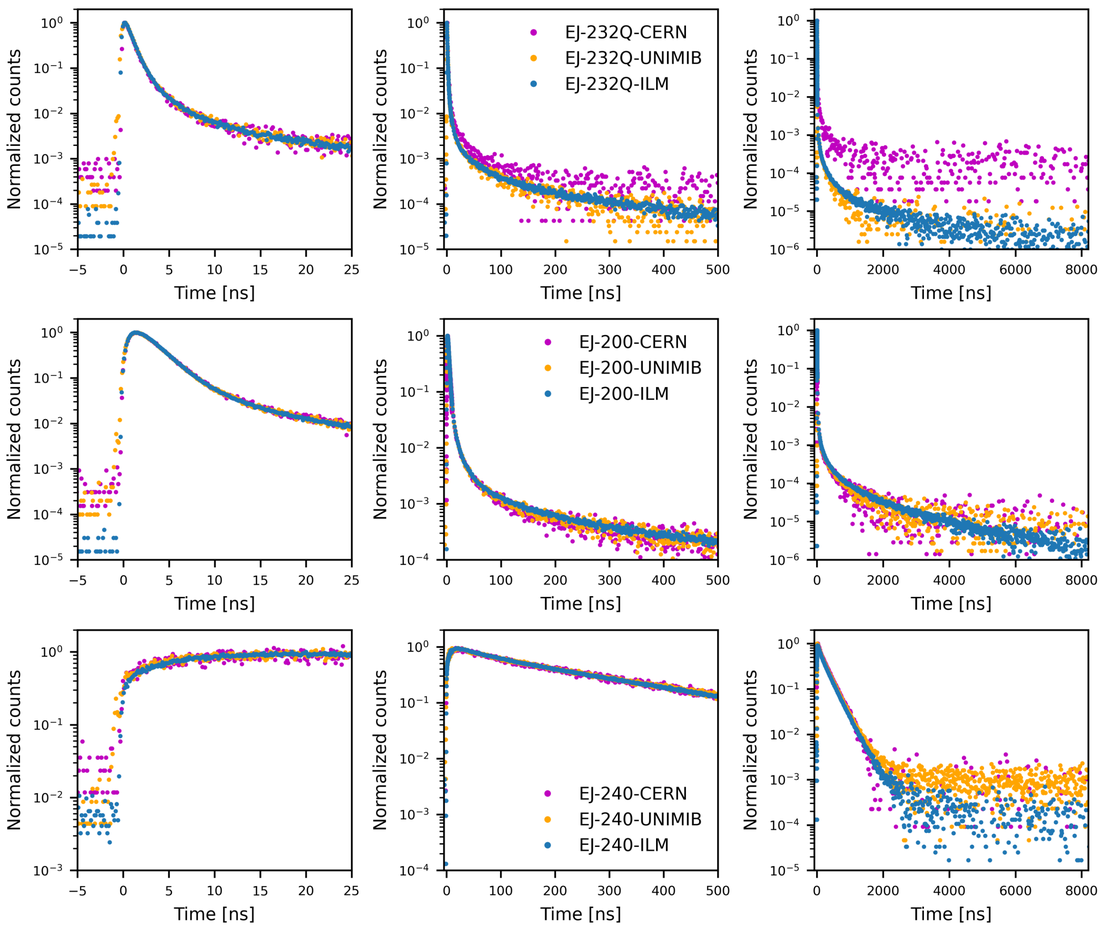
Introduction: Unlike single crystals, for which measurement methods are now well established, basic scintillation parameters such as scintillation yield are not yet standardized for nanoscintillators (NS) and nanocomposite scintillators.
Unlike optical excitation, scintillation mechanisms involve multiple energy relaxation steps, with characteristic distances significantly larger than nanoparticle sizes. Consequently, nanoparticles experience substantial charge and energy escape, and the overall scintillation process results from an interplay between the nanoscintillator and its host, which is typically a liquid or an organic solid. In this context, the medium can be considered as an ensemble comprising both the host and the NS.
Within the scope of Work Package 2 of DRD5 (https://drd5.web.cern.ch), this project aims to present a strategy to rationalize nanoscintillator measurements for the scintillation community. We also intend to establish a virtual laboratory based on four recognized European laboratories that have agreed to contribute to the development of a standardized measurement protocol.

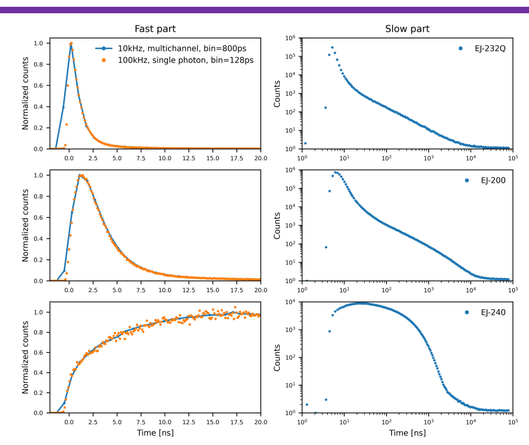
Selection of reference scintillators: As a first step, we aimed to cross-calibrate the fundamental properties of standard scintillators, namely radioluminescence spectra, timing characteristics, and scintillation yield. Since nanocrystals are generally dispersed either in plastics or in solvents, we selected representative reference materials for both configurations. Three plastic scintillators were chosen as reference standards and were graciously provided by Eljen Technology: EJ-200, EJ-240, and EJ-232Q-0.5%. For the liquid scintillator reference, we used Ultima Gold from Revvity.
To avoid unnecessary sample manipulation, particularly because some samples may contain toxic elements such as Pb or Cd, all samples have been — and will continue to be — measured directly in 6 mL Pico-Prias scintillation vials (Revvity). A standardized measurement geometry was defined as follows:
•**Plastic scintillators:** cylinders of 20 mm height and 12.5 mm diameter, with a fine diffusive finish
•**Liquid scintillator:** 2 mL filling volume in the scintillation vial.

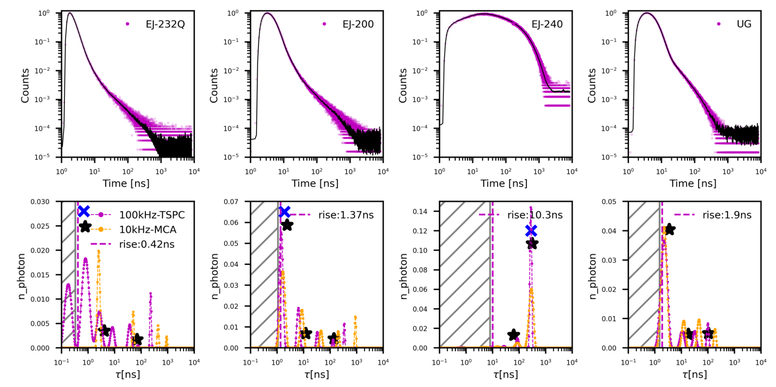
Timing performance was measured under X-ray excitation at CERN, UNIMIB, and ILM. A repetition rate of 100 kHz was used to enable measurements over a 10 µs time window. The data measured on 3 similar samples demonstrate the same results.



Scintillation decay time of the 3 standard plastics scintillators presented for 3 time scales.

Timing performance (slow part) By reducing the repetition rate to 10 kHz and using a multichannel analyzer (0.8 ns/bin), timing measurements under X-ray excitation can be extended up to 100 µs with sub-nanosecond resolution and a dynamic range of approximately six orders of magnitude. This enables the detection of possible the triplet emission in plastic scintillators. The fast component overlaps very well with the 100 kHz measurements, and progressive rebinning was applied to extend the dynamic range. It allows a more precise characterization of the slow component of the scintillation decay time.





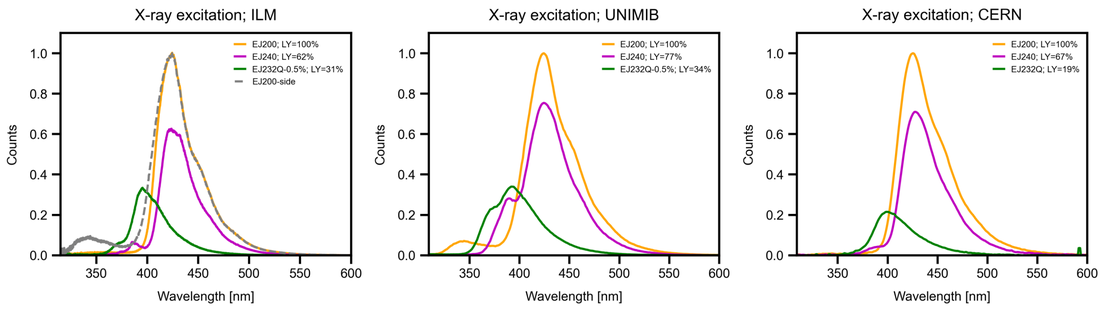
The blue crosses correspond to the values reported in the manufacturer's datasheet, while the black stars represent the parameters extracted from the fitting procedure applied to the CERN data, using three decay times and three rise times (two rise times for EJ240).

Analysis: Scintillation decay-time analysis is complex and typically dependent on a predefined number of exponentials.

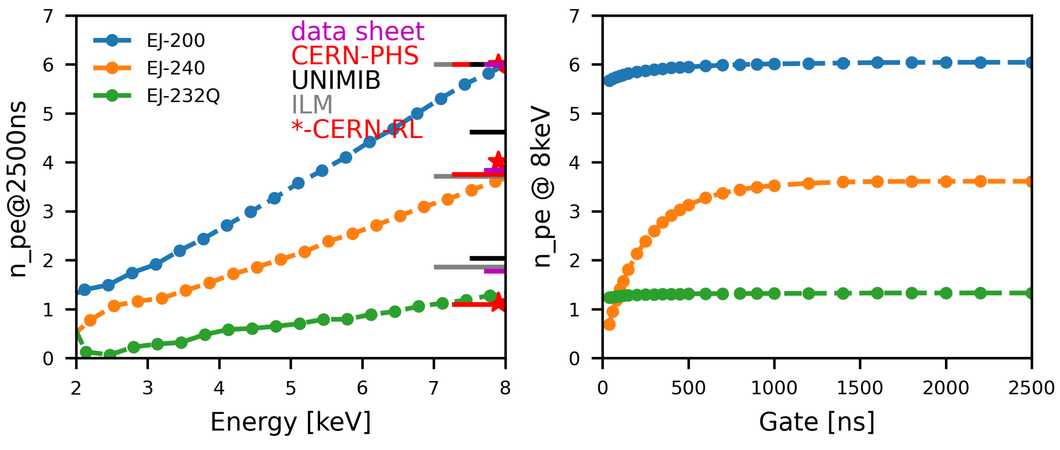
Here, we use an alternative approach (Laplace transform) with 500 predefined components, where only amplitudes are fitted, including a rise time and convolution with the IRF (laser, detector and X-ray tube).

A Tikhonov regularization stabilizes the algorithm and suppresses spurious components, yielding a photon-weighted lifetime distribution.

Radioluminescence, Non-proportionality and Light yield:
Radioluminescence was measured in "reflection" mode at UNIMIB, and at 90° with respect to the X-ray beam in Lyon and at CERN. The comparative yields were extracted and compared to other methods.



Comparison with other techniques and summary:
Pulsed height spectra (PHS) of plastics in vials were measured under excitation by a ^{137}Cs source, and the scintillation yield was deduced using the Compton edge approach. Compared to a standard 3 × 3 cube with all faces polished, a factor of 13 loss in light collection was observed (not shown here).
The Compton Triple-to-Double Coincidence Ratio (TDCR) method is an experimental technique developped at CEA. It enables measurement of the scintillation yield as a function of electron energy in the low-energy range (see Poster A. Perret @ this conference). The measurement at 8 keV as a function of the time window is consistent with the decay time measured in scintillators.
In contrast to yield estimations based on radioluminescence spectra—which measure emitted intensity under comparable excitation conditions—these 2 methods determine the number of detected photons per unit of deposited energy.
Taking EJ200 as the reference, all methods can be compared. Since the density and geometry of the plastic scintillators are similar, the relative yields are in good agreement.



Nanoscintillators: Measuring the scintillation yield of nanoparticles in liquid or solid media involves several critical factors, depending on the targeted information.

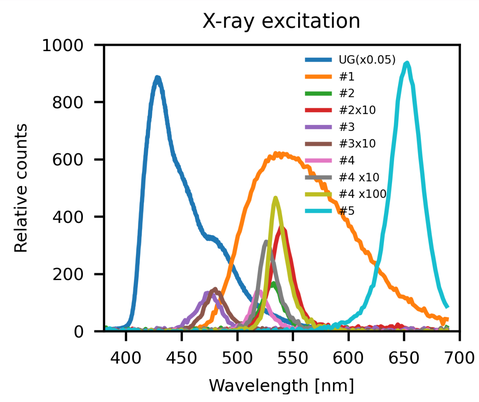
Radioluminescence intensity alone depends on multiple parameters, notably the concentration of the active material, the intrinsic scintillation of the host medium, and the potential interplay between them. Concentration influences X-ray absorption as well as the probability of secondary electron interactions with other nanoparticles. In cases of a small Stokes shift, it can also lead to self-absorption.

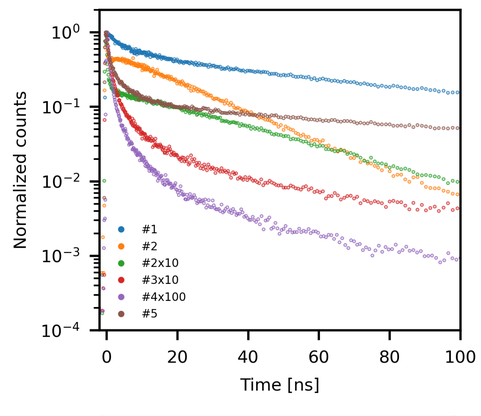
Most quantum dots are direct bandgap materials and therefore exhibit strong self-absorption. As shown in the graph, increasing the concentration by a factor of 10 leads to only a modest increase of the radioluminescence intensity even though the system remains in the weak X-ray absorption regime. A spectral shift is also observed.

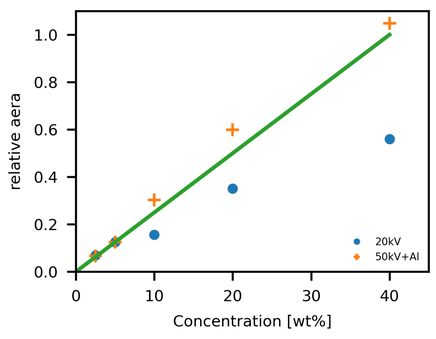
In the case of YAG:Ce, and more generally inorganic doped materials, self-absorption is negligible. By diluting a sample initially loaded at 40 wt%, we observe a clear effect of X-ray absorption saturation at low excitation energies (blue circles). At higher energies (orange crosses), a superlinear increase—attributed to secondary electron interactions—is first observed, followed by the onset of X-ray absorption saturation.

All these effects must be carefully considered when evaluating the scintillation efficiency of composite systems. From a materials optimization perspective, this light-yield measurement technique requires knowledge of the mass concentration to enable an accurate comparison of intrinsic performance, as displayed in the table above.

To evaluate performances and optimize the nanoscintillators themselves, their surface functionalization, and their interactions with supporting media, it is necessary to measure both the external and internal scintillation yields. The latter can be determined using the COMPTON TDCR experiment.







	composition	shape	Size (nm)	solvent	g/L	LY (%UG) (RL approach)	t@1/e (ns) (from I_{max})
#1	$Y_3Al_5O_{12}$:Ce	Sph.	8	water	420	7	21.6
#2	CdSe/CdZnS	Sph.	10	dodecane	5	0.5 (1; massx10)	7.8
#3	CdS/CdZnS	Sph.	10	dodecane	5	0.5 (0.4; massx10)	1.0
#4	$CsPbBr_3$	Cubes	10	dodecane	1	0.3 (0.7; massx10) (0.9; massx100)	0.8
#5	CdS/CdSe/CdS	Sph.	25	dodecane	16	3.6	1.5

Top (respec. middle): *Comparison of the RL spectra (respec. X-ray decay times) of NSs in solution (sample # refer to the table on the left).*
Bottom: *Evolution of the RL intensity as a function of the concentration of YAG:Ce NPs (#4) for two X-ray energies.*
Left: *Table of the studied NSs and their main measured scintillation characteristics.*

Conclusion: This working group is prepared to characterize samples of nanoscintillators in both solvents and polymer matrices provided by external laboratories. The objective is to move toward the standardization of measurement protocols. While scintillation decay measurements appear to be relatively robust, the determination of scintillation yield remains more challenging, in particular in non-homogeneous media. We believe that this strategy of standardization—based on consistent sample geometry and predefined measurement protocols—will benefit the nanoscintillator field.

Data base: This informal consortium, within the framework of the DRD5 research group, aims to establish a database of nanoscintillators, considered as nanocrystals in either solid or liquid media, evaluated under standardized geometry and experimental conditions. This work follows an open-source approach, and all data will be made available and regularly updated in accordance with a data management plan currently under development.

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