Electronic Supplementary Material (ESI) for Nanoscale.

Surface modification effect on contrast agent efficiency for X-rays based Spectral Photon-Counting Scanner / Luminescence Imaging: from fundamental study to in vivo proof of concept.

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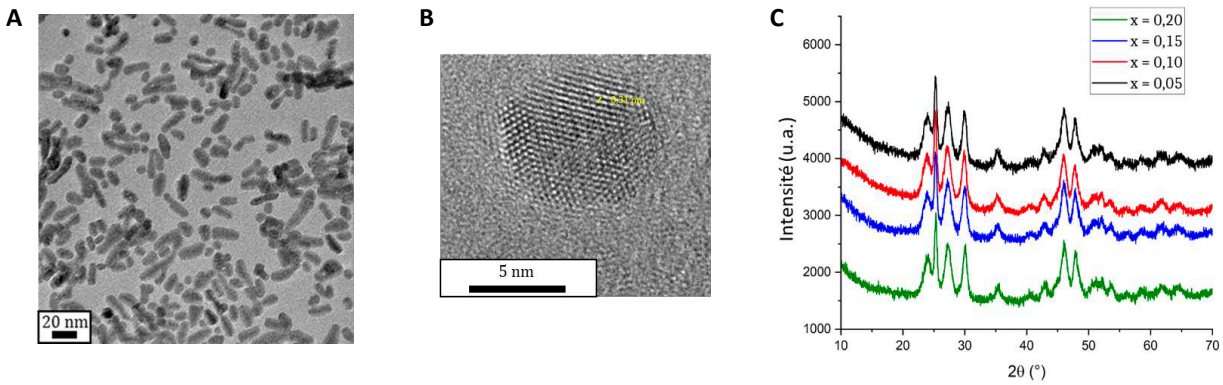


Fig. S1 Characterization of **NSc**. **A)** TEM image of $Gd_{0,90}Tb_{0,10}F_3$ composition. **B)** HRTEM of a nanoscintillator with the $Gd_{0,90}Tb_{0,10}F_3$ composition **C)** Measured XRPD pattern of various composition (x is the molar ratio of Terbium) corresponding to the orthorhombic phase (*Pnma* space group) of GdF_3.

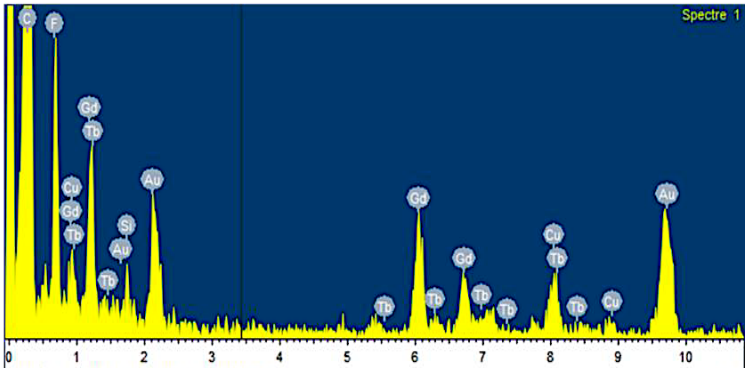
Table S1 Theoretical and measured Tb^{3+}/Gd^{3+} ratio for the various NCs compositions

Composition	*Theoretical*	*ICP*
$Gd_{0.95}Tb_{0.05}F_3$	0,05	0,06
$Gd_{0.90}Tb_{0.10}F_3$	0,10	0,11
$Gd_{0.85}Tb_{0.15}F_3$	0,15	0,18
$Gd_{0.80}Tb_{0.20}F_3$	0,25	0,23

Table S2 DLS measurements (hydrodynamic diameters *Dh* and Polydispersity index PDI) on the various composition in water (pH=6) and zeta potential (ZP) measurements.

Composition	*Dh (nm)*	*PDI*	*ZP (mV)*
$Gd_{0.95}Tb_{0.05}F_3$	16 ± 3	0,09	48 ± 2 mV
$Gd_{0.90}Tb_{0.10}F_3$	13 ± 3	0,11	45 ± 2 mV
$Gd_{0.85}Tb_{0.15}F_3$	13 ± 3	0,10	44 ± 2 mV
$Gd_{0.80}Tb_{0.20}F_3$	16 ± 3	0,11	49 ± 2 mV

A



B

Elements	Atomic percentages
Gd	39,21
Tb	3.74
F	57.11

Fig. S2 EDX analysis of $Gd_{0.90}Tb_{0.10}F_3$ composition **A**) Spectrum of the NSc **B)** Analysis results.

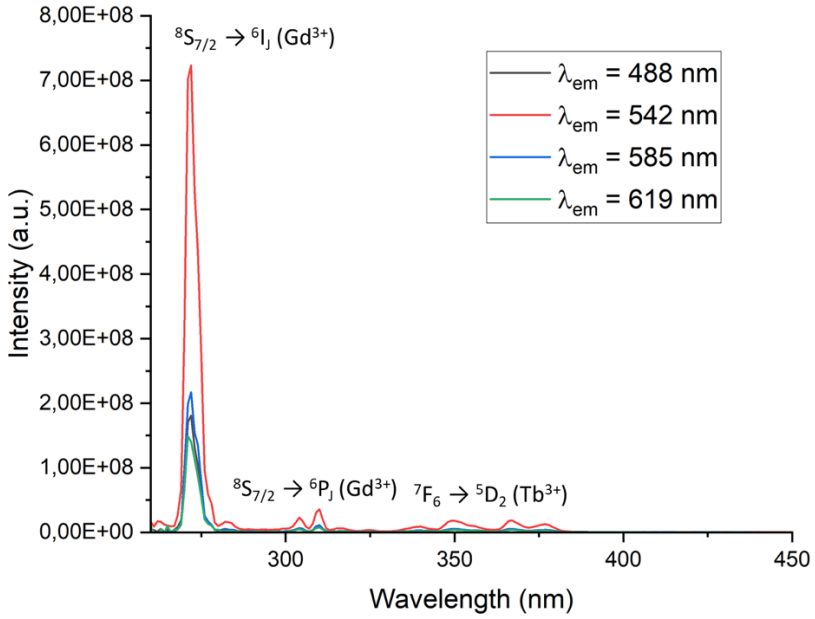


Fig. S3 Excitation spectra of the $Gd_{0.90}Tb_{0.10}F_3$ NSc.

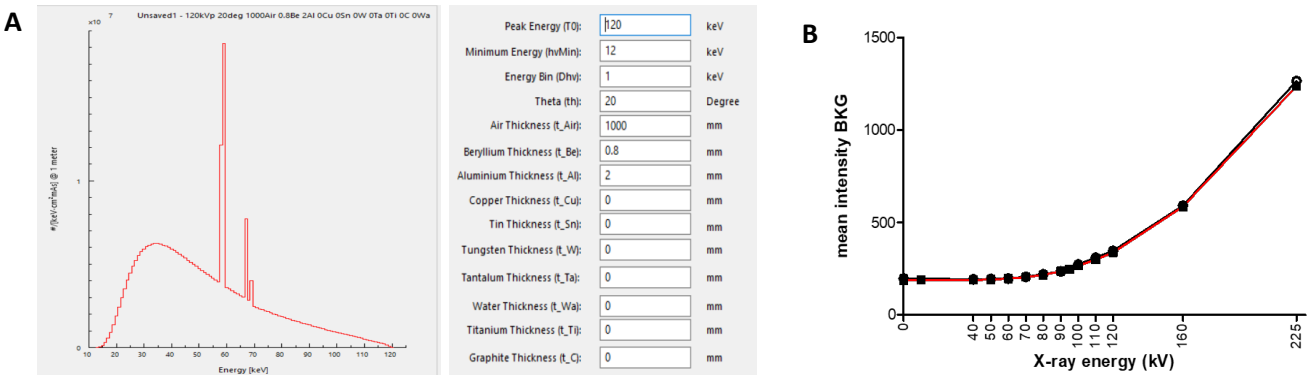


Fig S4. **A)** Spectrum of X-rays with peak energy of 120 keV (obtained using SpekCalc software (PMID: 19724100). Right : Parameters used for calculation. **B)** Increase in the intensity of background with increasing X-ray energy.

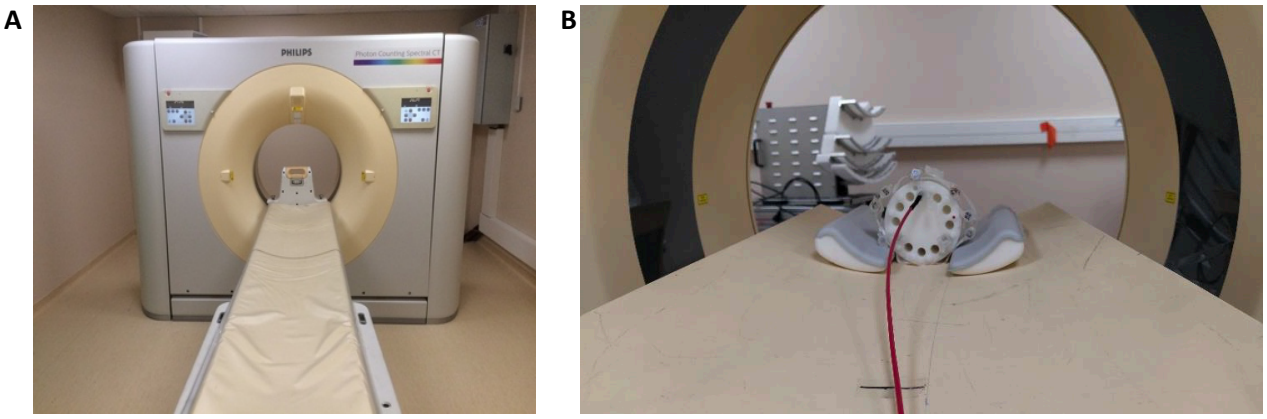


Fig S5. **A)** General picture of the Spectral photo counting scanner CT. **B)** Picture of the experimental setup with optical fiber immersed in a colloidal suspension within an Eppendorf tube. The fiber is linked to a spectrophotometer for luminescence measurements.

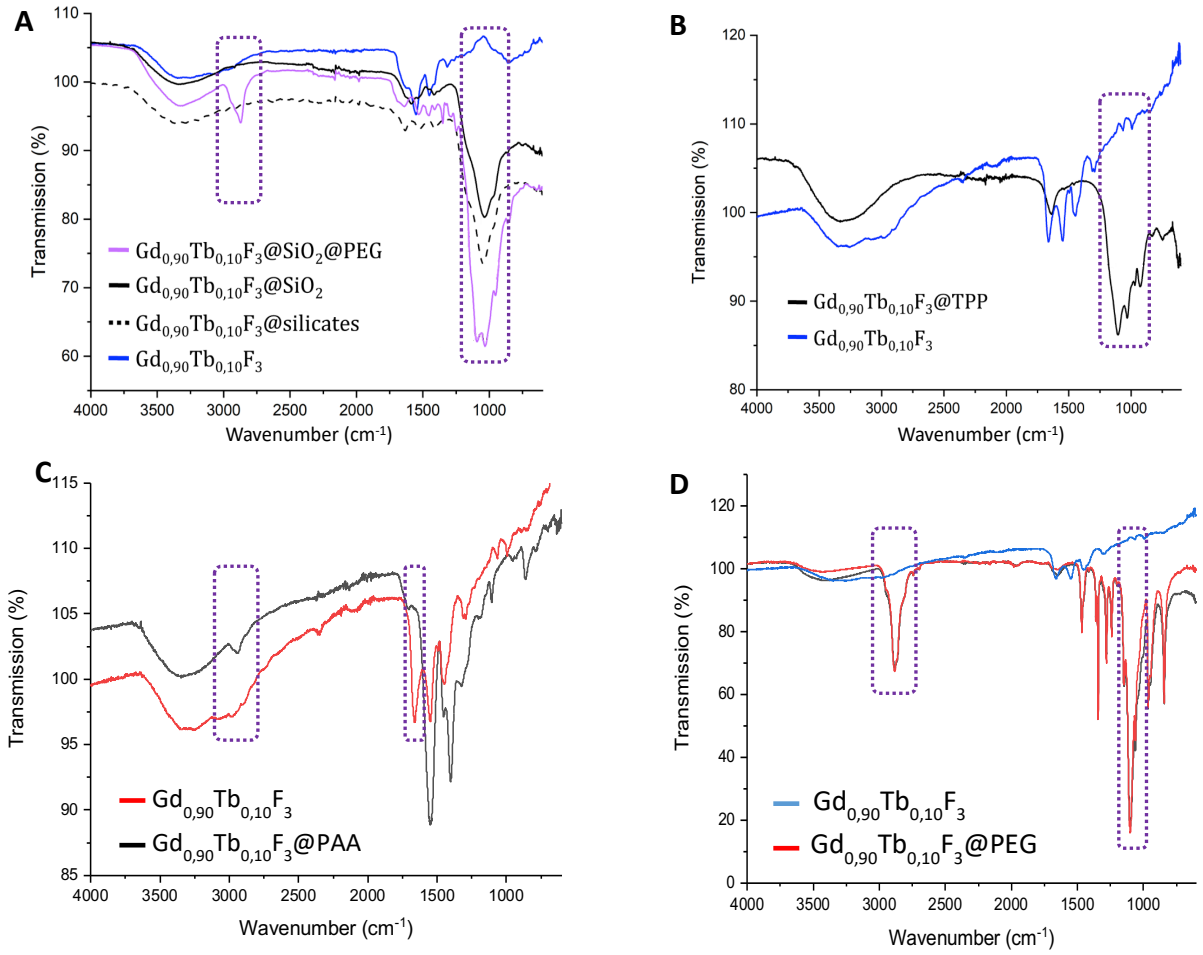


Fig S6. FTIR spectra of $Gd_{0.90}Tb_{0.10}F_3$ after surface modification, main bands are circled in purple boxes **A)** FTIR monitoring of NSc surface modification with SiO_2@PEG layer at each steps of the process. **B)** Spetra of the NSc before (blue line) and after (black line) grafting of TPP molecules. **C)** Spectra of the NSc before (red line) and after (black line) grafting of PAA polymer. **D)** Spectra of the NSc before (blue line) and after (red line) grafting of PEG polymer.

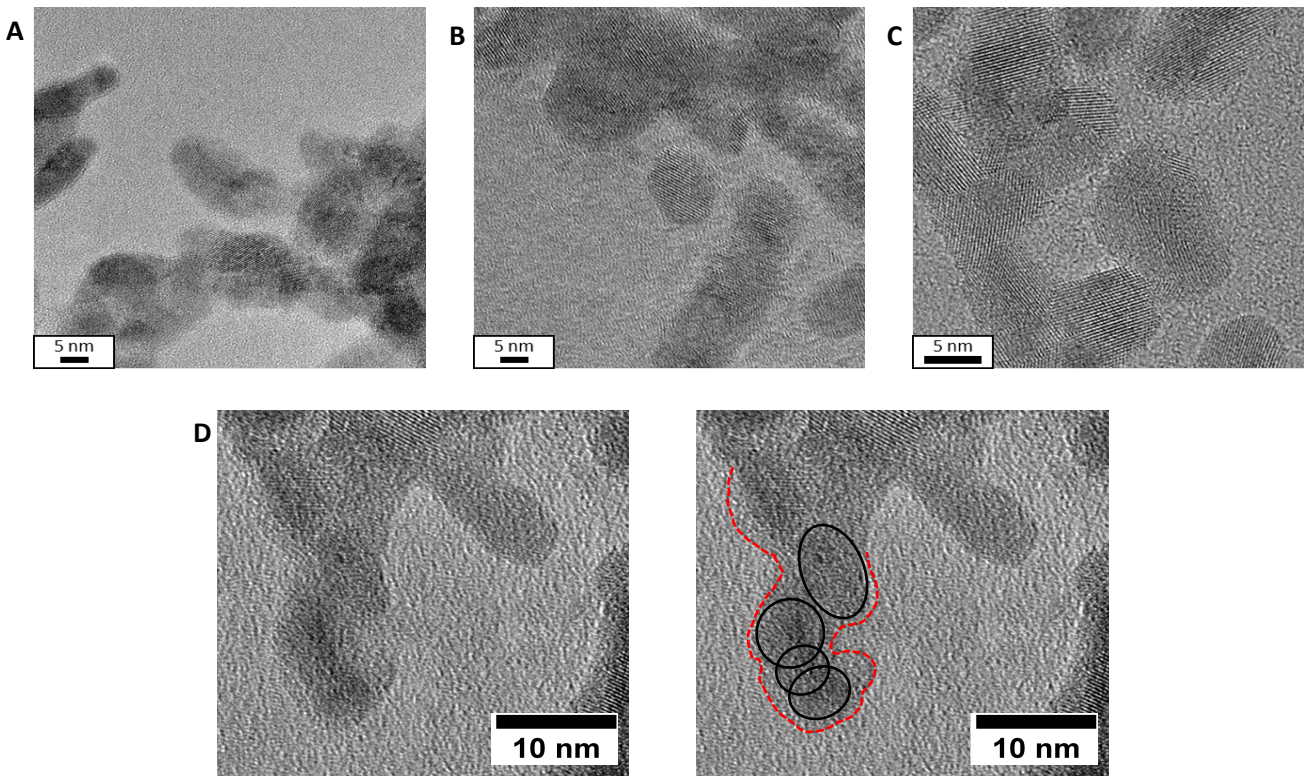


Fig S7. HRTEM of $Gd_{0.90}Tb_{0.10}F_3$ after surface modification with various ligands. **A)** $Gd_{0.90}Tb_{0.10}F_3$@TPP. **B)** $Gd_{0.90}Tb_{0.10}F_3$@PAA. **C)** $Gd_{0.90}Tb_{0.10}F_3$@PEG. D) $Gd_{0.90}Tb_{0.10}F_3$@SiO_2@PEG. Picture on the left as observed, picture on the right same area with schematic representation of the nanoparticles (black line) and the silica layer (red line).

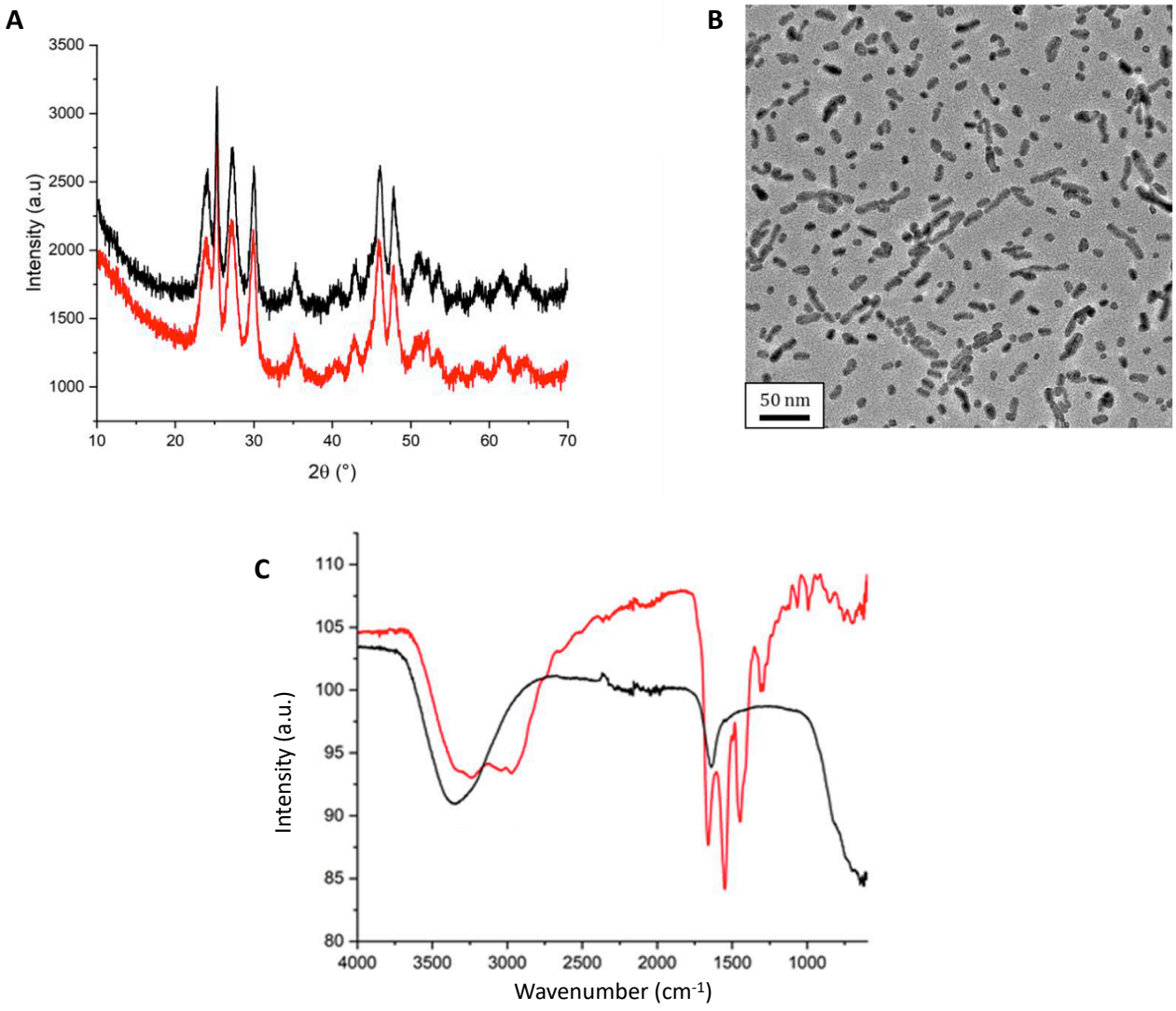


Fig S8. **A)** PXRD of $Gd_{0.90}Tb_{0.10}F_3$ before (red line) and after (black line) cleaning treatment of the remaining 2-pyrolidinone. **B)** TEM picture of cleaned $Gd_{0.90}Tb_{0.10}F_3$. **C)** FTIR spectra of $Gd_{0.90}Tb_{0.10}F_3$ before (red line) and after (black line) cleaning treatment of the remaining 2-pyrolidinone.

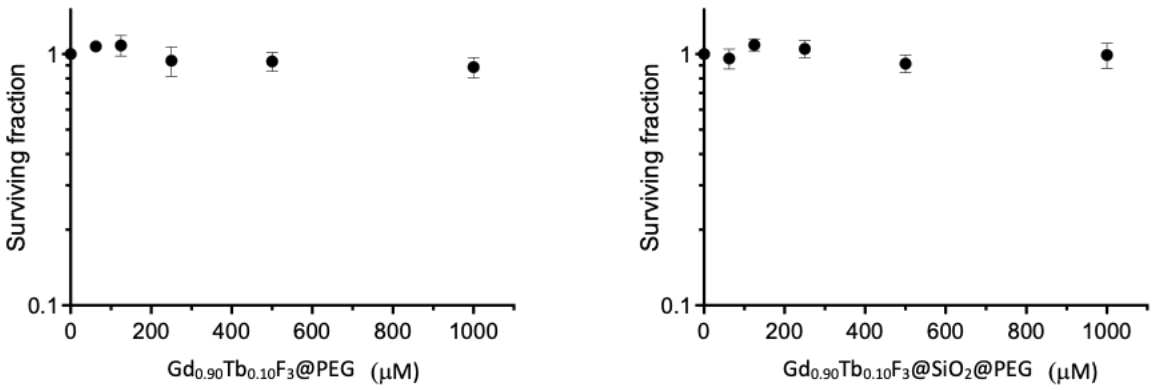


Fig S9. Surviving fraction of clonogenic MDA-MB-231 breast cancer cells upon treatment with the range of nanohybrids concentration for 24 h. Mean of 3 biological repeats ± SD.