Supplementary Information (SI) for Journal of Materials Chemistry C.

Supporting Information of

Employing Eu,Tb-doped HfO_2 nanocrystals as built-in cryogenic luminescent nanothermometer for commercial coated conductors

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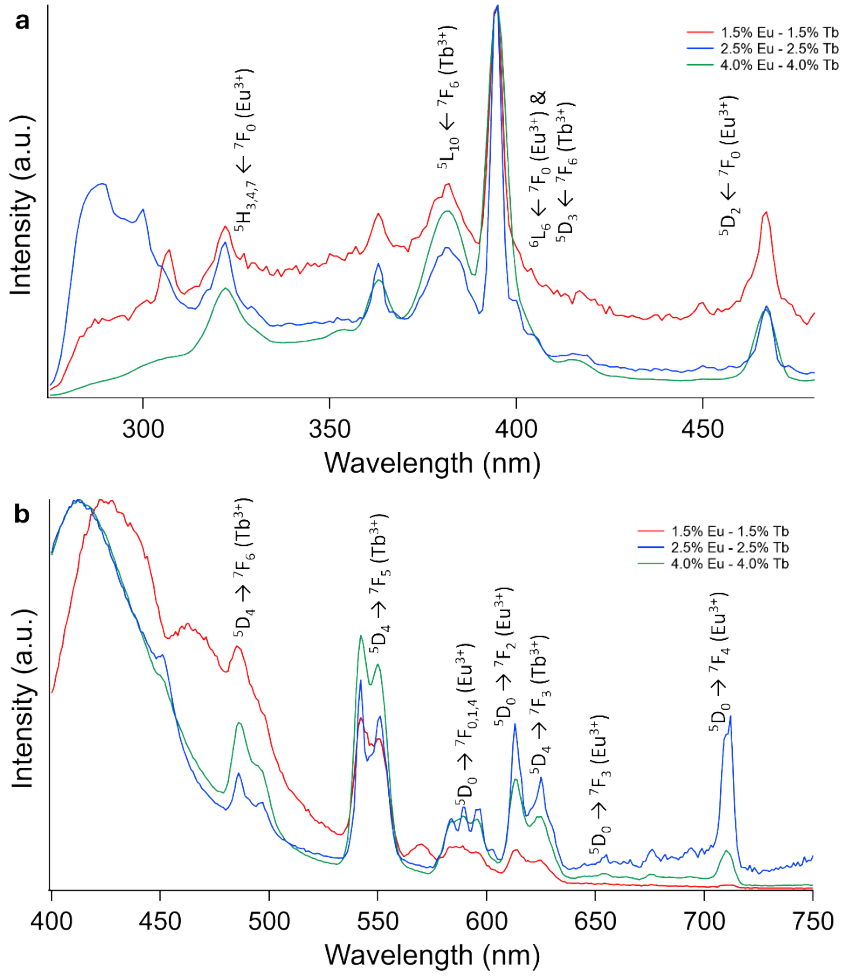

Figure S1. Comparison of normalized photoluminescence spectra of HfO_2:Eu^{3+},Tb^{3+} NCs with different doping percentages. Red: HfO_2:1.5%Eu^{3+},1.5%Tb^{3+}, blue: HfO_2:2.5%Eu^{3+},2.5%Tb^{3+} and green:

HfO_2:4.0%Eu^{3+},4.0%Tb^{3+} NCs. (a) Excitation spectrum (λ_{em} = 613 nm), and (b) emission spectrum (λ_{ex} = 285 nm). Comments on the spectral features are found in table S1.

Table S1. Overview of the screening process to determine the total doping percentage.

Doping percentage Eu^{3+}	Doping percentage Tb^{3+}	Description of the emission spectrum
1.50	1.50	As starting material, nanoparticles doped with 1.5% of each lanthanide were synthesized. This total doping percentage was not enough to produce f-f transitions peak with a good intensity compared to the broad band that is present around 400 - 475 nm.
2.50	2.50	The intensities of the f-f transition peaks to the intensity of the broad band was much better compared to 3% total doping. Moreover, compared to 3% total doping, there is no substantial reduction in decay time for 5% total doping percentage.
4.00	4.00	The intensities of the f-f transition peaks to the intensity of the broad band is more or less same as the 5% total doping. Therefore, a final total doping percentage of 5% was chosen.

Table S2. Overview of the screening process to determine the ratio of the Eu^{3+}/Tb^{3+} ratio.

Doping percentage Eu^{3+}	Doping percentage Tb^{3+}	Description of the emission spectrum
2.50	2.50	As determined in the previous table, a total of 5% doping is best for this application. However, the Eu:Tb ratio was not optimal: the intensity of the Eu peaks were not strong enough compared to the Tb peaks.
3.75	1.25	The ratio of the Eu peaks compared to the Tb peaks was better at room temperature for this sample, however in the cryogenic region this doping ratio resulted in a thermometry system with poor S_r values.
4.50	0.50	This ratio of the Eu peaks to the Tb peaks was excellent in this system. Furthermore, this doping ratio in the cryogenic region yielded a thermometry system with good S_r values. Therefore, these doping percentages were chosen for this system and application.

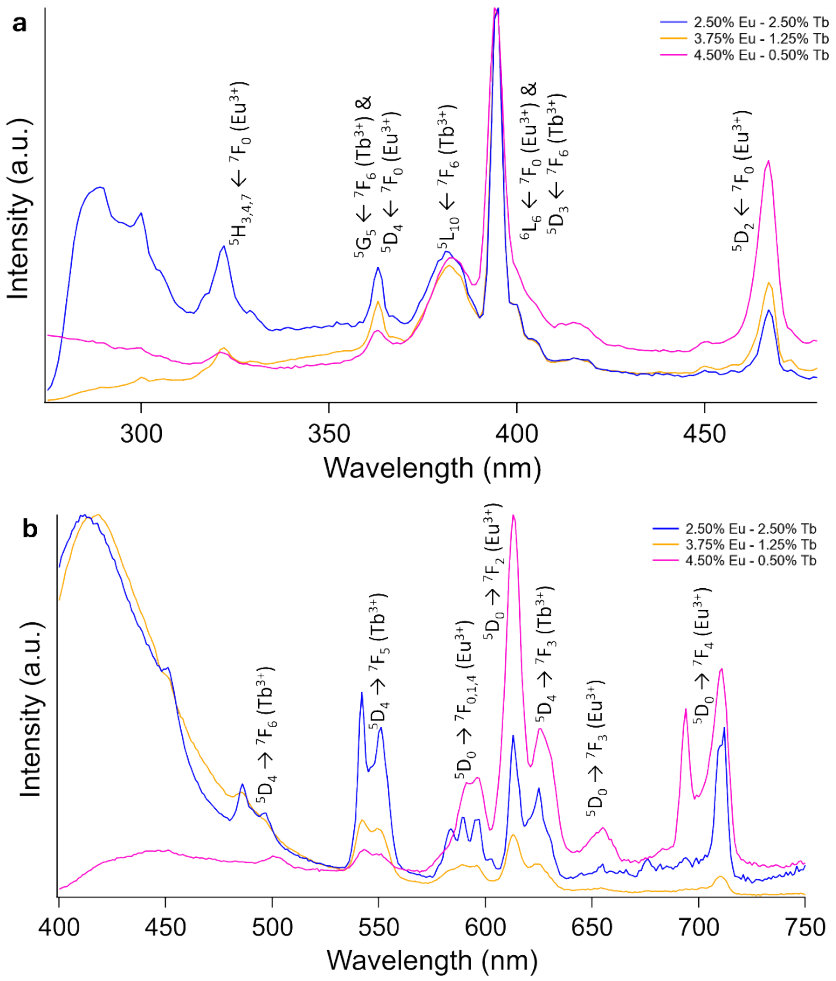

Figure S2. Comparison of normalized photoluminescence spectra of HfO_2:Eu^{3+},Tb^{3+} NCs with a total of 5% lanthanide doping, but different doping ratios. Blue: HfO_2:2.5%Eu^{3+},2.5%Tb^{3+}, orange: HfO_2:3.75%Eu^{3+},1.25%Tb^{3+} and pink: HfO_2:4.5%Eu^{3+},0.5%Tb^{3+} NCs. (a) Excitation spectrum (λ_{em} = 613 nm)), and (b) emission spectrum (λ_{ex} = 285 nm). Comments on the spectral features are found in table S2.

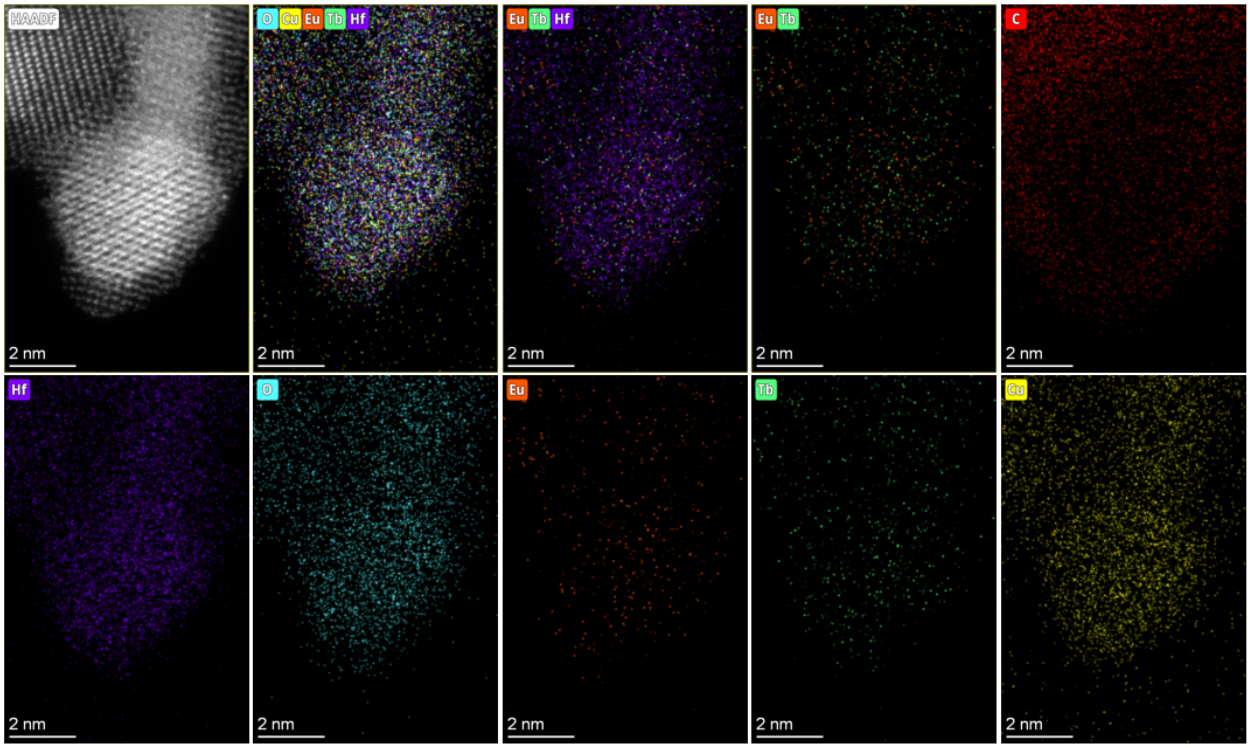

Figure S3. High-angle annular dark-field (HAADF)-scanning transmission electron microscopy (STEM) with corresponding energy-dispersive X-ray spectroscopy (EDX) compositional and element maps.

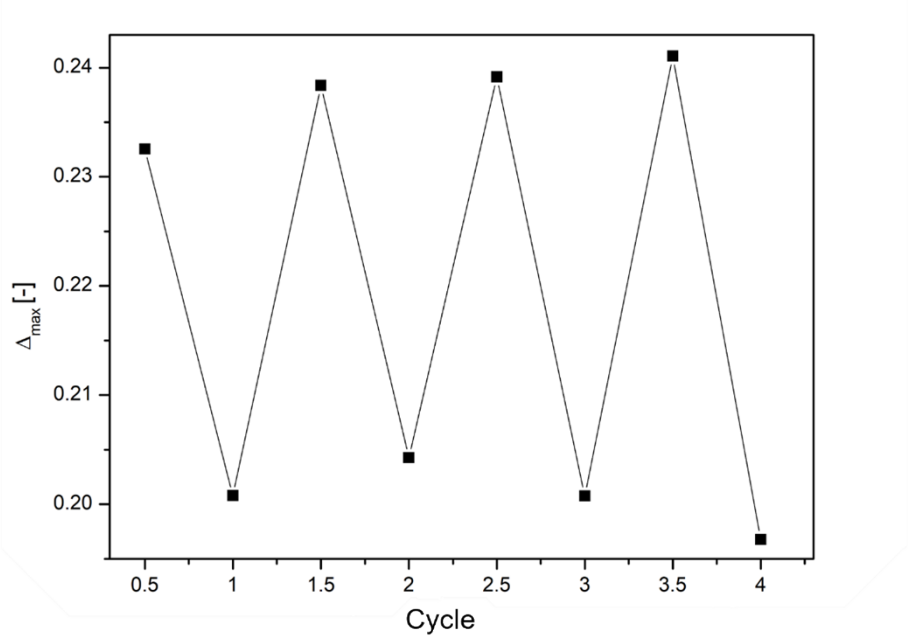

Figure S4. Cycle test of HfO_2:4.5%Eu^{3+},0.5%Tb^{3+} nanocrystals, measured between 40 K and 100 K with a step size of 10 K. A repeatability of 98% was determined using eq. 4.

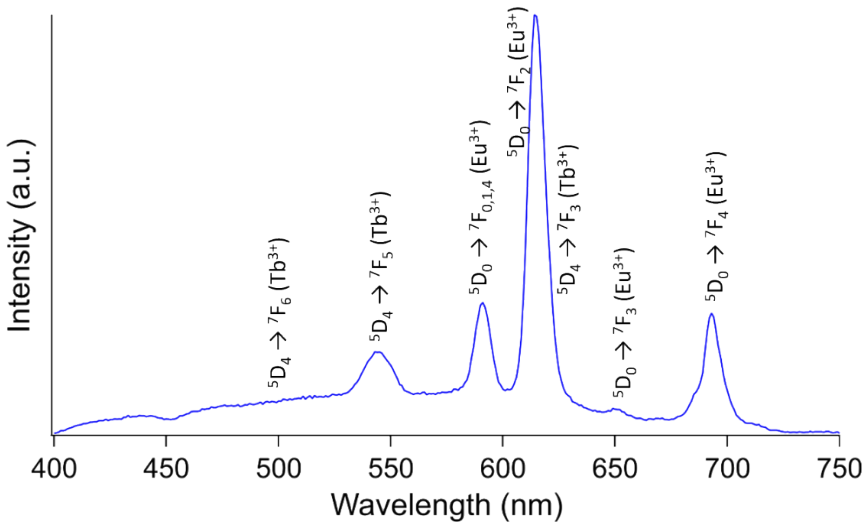

Figure S5. Emission spectrum of HfO_2:4.5%Eu^{3+},0.5%Tb^{3+} measured at RT as a thin film. The excitation wavelength is 250 nm.

The data of the film recorded at RT clearly shows the Tb^{3+} 540 nm peak in a similar Eu^{3+}/Tb^{3+} ratio as in the powder. The peaks look a bit different and are embedded on a broad band, but that is due to it being a film.

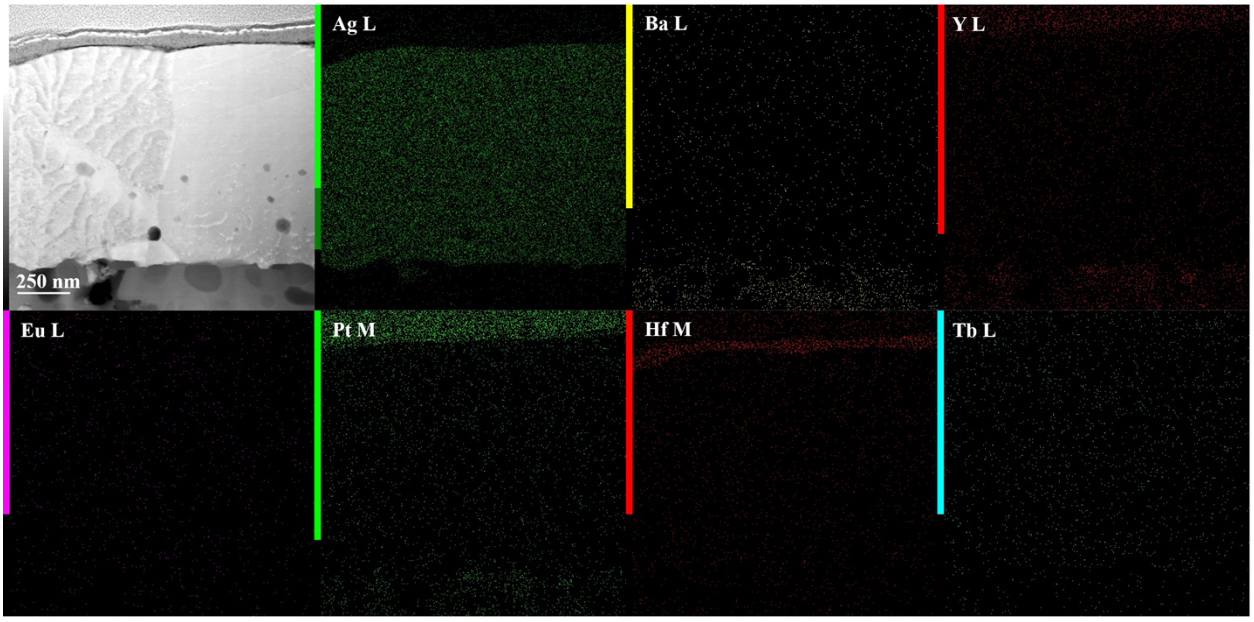

Figure S6. EDX compositional and element maps of a commercial YBCO tape with 60±10 nm HfO_2:4.5%Eu^{3+},0.5%Tb^{3+} nanocrystals on the top of Ag protective layer at low magnification.