**Mesoscale Architecture of RE-doped $HfO_2$ Nanophosphors: Does Local Density Matter?**

Laura Henz[1], Joshua Galvis Melo[1], Alessandro Lauria[1]

[1]*Laboratory for Multifunctional Materials, Swiss Federal Institute of Technology (ETH), Zurich, Switzerland*

alessandro.lauria@mat.ethz.ch

Luminescent nanoparticles have recently gained significant attention within the scintillator research community, thanks to the promise for expanded design flexibility regarding more complex geometries and easier manufacturing. Indeed, various architectures based on nanoparticles - such as aerogels, optical polycrystalline ceramics, and embedded plastic nanocomposites - seem to allow the combination of versatile forming with the high performance of inorganic scintillators. While larger-scale scintillators become accessible through these methods, the local density of the material remains lower than that of bulk single crystals, which potentially results in a weaker interaction between the material and ionizing radiation. On the other hand, the discrete spatial organization of single crystalline particles may hinder light transport following the radioluminescence process due to phenomena such as scattering or self-absorption. Also in the field of nanomedicine, like in X-ray Induced Photodynamic Therapy (X-PDT), this morphological aspect is critical. Nanoparticles frequently reorganize in an inhomogeneous manner - for instance, following internalization into cellular environments - which makes it difficult to predict their overall behavior within biological environments.

To elucidate the role of local density on the X-ray stimulated phenomena in these NP-based materials, it is essential to precisely control their short-to-mid-range structural organization. Recent studies suggest that radioluminescence (RL) efficiency is diminished if secondary electrons, generated within nanophosphors upon energy deposition, are not intercepted by neighboring particles in the immediate vicinity [1]. Accordingly, a perfectly monodisperse distribution might optimize visible light transmittance while suppressing the stopping power. The lack of active material in the gaps between small particles facilitates the 'escape' of secondary electrons, thereby failing to feed the scintillation process.

While recent works have attempted to correlate the local density of nanoparticles with overall performance, an underlying general rationale is not yet fully understood.[2] In the present work, we compare the radiation attenuation of RE-doped (Tb, Ce) $HfO_2$ ultrasmall nanoparticles, obtained by the solvothermal route, and organized in samples with increasing local density distributions. These configurations include: singly dispersed dispersions, partially agglomerated (destabilized) suspensions, aerogels, and dense particle assemblies (films). We evaluate the relationship between the NPs submicron local density and X-ray retention, light transmittance, as well as the luminescence stimulated by either ultraviolet (UV) light or ionizing radiation.

1. V. Vistovskyy et al., "Modeling of X-ray excited luminescence intensity dependence on the nanoparticle size", *Radiation Measurements*, **90**, 174 (2016).

2. B.T. Diroll et al., "Effects of Quantum Dot Loading on the Radioluminescence Efficiency in Quantum-Dot-Embedded Composites," *Nano Letters*, **25**, 16733 (2025).