Cerenkov Luminescence Signal Enhancement using Hyperbolic Metamaterials as a Multi-modal Imaging Probe

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Cerenkov Luminescence (CL) is a phenomenon observed when a charged particle is emitted at a phase velocity faster than the speed of light in a chosen dielectric medium[1-3]. CL is inherently multi-modal in that it uses the same FDA-approved radiotracers as Positron Emission Tomography (PET), yet with faster imaging speeds, lower service costs, higher resolution, and no incident light source[1-3]. CL is emitted as a relatively weak blue light, which so far limits clinical translation due to low tissue penetration. However, a CL contrast agent could enhance the CL signal without increasing the dose of radioactivity for patients[2-3]. Current work focuses on designing down-converting materials to convert CL into red-light emissions to increase penetration and to reduce tissue absorption[2]. Our objective is to couple this approach with hyperbolic metamaterials (HMMs), or subwavelength structures, to directly enhance the CL photon yield by reducing the energy threshold needed to generate CL[3]. HMMs consisting of silver (metal) and alumina (dielectric) alternating layers were fabricated using chemical vapor deposition and HMMs with holes in the metamaterial were fabricated as byproducts of Janus particle studies. The HMMs were analyzed using an optical microscope at 20x, 50x, and 100x magnification. Eu_2O_3 nanoparticles (NPs) were characterized using Transmission Electron Microscopy (TEM), Dynamic Light Scattering (DLS), and fluorescence to confirm optical properties needed as a downconverter for CL. The Eu_2O_3 NPs were radiolabeled with Yttrium-90 (^{90}Y) by mixing 1mL of Eu_2O_3 NPs (10mg/mL) sonicated in 10% methanol (aqueous) with 51μL of [^{90}Y]YCl_3 solution (7.4MBq) at a temperature of 95°C for up to an hour. Time points were taken every 20 minutes and analyzed using radio-TLC. To analyze the effect of HMMs on light propagation, 100μL of Eu_2O_3 NPs (10mg/mL) in 10% methanol was spun onto the HMMs using a Cytospin, and then imaged using the small animal imaging system (IVIS spectrum). TEM images displayed sizes of Eu_2O_3 NPs ranging from 117.96nm to 250.94nm. Scanning Transmission Electron Microscopy (STEM) Energy-Dispersive x-ray Spectroscopy (EDS) confirms the elemental composition of Eu_2O_3 NPs to be europium and oxygen. DLS results of Eu_2O_3 NPs demonstrated a hydrodynamic size of 189.2nm in 10% methanol (10mg/mL). The Eu_2O_3 NPs were excited at 270nm with two emission peaks at 620nm (characteristic of Eu^{3+} oxidation state) and 700nm. The radio-TLC demonstrated the [^{90}Y]YCl_3 traveling further from the origin than the [^{90}Y]Y- Eu_2O_3 NP complex at about half that distance, proving the yttrium-90 had adsorbed to the Eu_2O_3 NPs' surface after thermal reacting for more than 20 minutes. Compared to a control of Eu_2O_3 NPs on a blank microscope slide, the HMMs showed an average increase in Eu_2O_3 NPs luminescence signal from 3.49×10^8 to 5.94×10^8 total radiancy. So far, these promising findings show the HMMs can increase the photon yield of Eu_2O_3 NP luminescence by up to 70%, supporting our hypothesis that HMMs can be used to enhance CL using the [^{90}Y]Y- Eu_2O_3 NP complexes in further studies.

References:

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