

Internationally, pH stands as a routinely measured chemical parameter, with glass pH electrodes serving as a widely acknowledged standard for their reliability, precision, and comprehensive pH range coverage. Despite their advantages, such as cost-effectiveness and prompt response, glass electrodes possess limitations like size, inflexibility, and vulnerability to external fields, constraining their applicability in specific contexts. Optical methods emerge as compelling alternatives to tackle pH measurement challenges, particularly at nanoscales and for imaging pH distribution.

The shortcomings of glass electrodes, encompassing fragility and the requirement for storage solutions, have spurred exploration into alternative optical pH sensing devices over recent decades. FRET-based sensors, particularly those harnessing upconverting nanoparticles (UCNPs), demonstrate potential in gauging local variations in critical biological parameters. UCNPs, emitting higher-energy photons upon exposure to lower-energy photons, offer advantages including resistance to photobleaching and background-free detection. The distinctive multi-colour emission of UCNPs facilitates ratiometric sensing, mitigating errors linked to variations in excitation intensity or nanoparticle concentration, making them a robust and accurate choice for diverse sensing applications.

However, despite FRET being a pivotal process in emissions, the potential photon reabsorption (PR) effect in upconverting nanoparticles poses a challenge. PR transpires when a photon emitted by one UCNP is absorbed by a dye on another nanoparticle, resulting in diminished emission and obscuring the authentic FRET response. This challenge arises due to the random distribution of emitting ions throughout the UCNP volume, with only a fraction near the surface participating in FRET. Many current sensors overlook this complexity, relying on less-sensitive reabsorption processes.

An encouraging solution involves employing core-shell structures, situating all donor ions in the shell, potentially enhancing FRET-based sensing while minimizing PR. The primary project objective is to comprehend the energy transfer mechanisms in core@shell sensors. By exploring different architectures and concentrations of codopant ions, a meticulous analysis of potential energy transfer routes will be conducted. The objective is to enhance the sensitivity of FRET-based pH sensors by reducing the simultaneous occurrence of the PR process. Positioning all donor ions within the FRET distance aims to ensure the complete transfer of energy to the dye. The hypothesis posited is that, with an appropriate core-shell structure, possibly housing all donor ions within the shell, FRET efficiency can be significantly enhanced, and the PR process minimized. Core@shell structures not only enhance FRET-based sensor efficiency but also unlock multifunctionality, revolutionizing nanomaterial design.

The project's secondary aim is to develop multisensor nanoparticles for local pH and temperature measurement or two different parameters simultaneously. Two approaches involve incorporating optically active ions as donors for distinct dyes, enabling the simultaneous observation of two emissions. The second scenario targets a dual-sensing multisensor responding to temperature and pH, utilizing a core@shell structure with temperature-responsive ions in the core and emission-compatible ions in the shell. This strategy holds promise for practical applications and advances fundamental understanding in multisensor design.