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### **ACCEPTED MANUSCRIPT**

# A novel "modularized" optical sensor for pH monitoring in biological matrixes

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**Abstract:** A novel core-shell structure optical pH sensor is developed with upconversion nanoparticles (UCNPs) serving as the core and silica as the shell, followed by grafting bovineserumalbumin (BSA) as another shell via glutaraldehyde cross-linking. The obtained core-shell-shell structure is shortly termed as UCNPs@SiO<sub>2</sub>@BSA, and its surface provides a platform for loading various pH sensitive dyes, which are alike "modules" to make it feasible for measuring pHs within different pH ranges by simply regulating the type of dyes. Generally, a single pH sensitive dye is adopted to respond within a certain pH range. This study employs bromothymol blue (BTB) and rhodamine B (RhB) to facilitate their responses to pH variations within two ranges, i.e., pH 5.99-8.09 and pH 4.98-6.40, respectively, with detection by ratio-fluorescence protocol. The core-shell-shell structure offers superior sensitivity, which is tens of times more sensitive than those achieved by ratio-fluorescence approaches based on various nanostructures, and favorable stability is achieved in high ionic strength medium. In addition, this sensor exhibits superior photostability under continuous excitation at 980 nm. Thanks to the near infrared excitation in the core-shell-shell structure, it effectively avoids the self-fluorescence

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