Announcing the Final Examination of Swetha Barkam for the degree of Doctor of Philosophy

Title: THE STUDY OF PHYSIOCHEMICAL PROPERTIES OF CERIUM OXIDE NANOPARTICLES AND ITS APPLICATION IN BIOSENSORS

Biosensors continue to get smaller and faster with the advancement in nanotechnology through the use of nanomaterials to achieve high sensitivity and selectivity. However, the continued reliance on biomolecules or enzymes in the biosensor assembly poses the problem of reproducibility, storage and complexity. The dissertation research address this challenges by investigating the physiochemical properties of nanoparticles to understand its interaction with biological entities and develop enzyme free biosensors. In this study, we have demonstrated a novel strategy to integrate cerium oxide nanoparticles (CNPs) as an efficient transducer through rigorous screening for developing enzyme/label free biosensors for detecting analytes such as dopamine associated with neurodegenerative diseases and limonin for fruit quality management. CNPs have been proven to exhibit antioxidant properties attributed to its dynamic change in surface oxidation states (Ce4+ to Ce3+ and vice versa) mediated at the oxygen vacancies on the surface of CNPs. It is well-established that nanoparticles are resourceful novel materials with a plethora of applications in the field of nanomedicine.

In one of the studies, the effects of different anions in the precursor of the cerium salts used for synthesizing CNPs using the same synthesis method, were extensively studies. It has been demonstrated that the physicochemical properties such as dispersion stability, hydrodynamic size, and the signature surface chemistry, antioxidant catalytic activity, oxidation potentials of different CNPs have been significantly altered with the change of anions in the precursor salts. The increased antioxidant property of CNPs prepared using the precursor salts containing NO3¯ and Cl¯ ions have been extensively studies using in-situ UV-visible spectroscopy which reveal that the change in oxidation potentials of CNPs with the change in concentration of anions. Thus, this work demonstrated that the physicochemical and antioxidant properties of CNPs can be tuned by anions of the precursor.

In another study, CNPs have been immobilized on highly ordered polymer nanopillars to develop an optical sensor for dopamine. Dopamine, is one of the main neurotransmitters which plays a significant role in central nervous system and its deficiency leads to neurological disorders such as Parkinson’s disease, schizophrenia etc. Current biosensors in the literature use invasive detection techniques and lacks sensitivity for physiological clinical relevant concentrations dopamine. The interaction between CNPs and dopamine have been extensively studied using UV-visible spectro-electrochemical studies to achieve the right surface chemistry (35%~70% Ce4+). The sensor exhibits high sensitivity (1fM detection in simulated body fluid), high selectivity (in acetic acid, sheep plasma) and increased robustness with several cycles of usage.

In the final study, CNPs integrated in silk fibroin (SF) polymer electrospun nanofibers incorporated on an organic electrochemical transistor platform, is used to develop a limonin sensor. It has been established that the concentration of limonin in citric fruit predicts the quality in terms of bitter taste from the HLB bacteria infected fruits. A unique in-situ electrospinning set-up using drum as collector was used to develop SF (extracted from cocoon) nanofibers used as CNP (synthesized in-situ in fibers) transducer carrier, both of which have a specific interaction with limonin. This novel biosensor has exhibited high sensitivity (100nM in PBS) and selectivity (citric acid, sugar etc) with improved robustness in terms of reuse.

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