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Title of thesis: Dynamic Studies on Thin Film based Sensors for Biomedical Applications

ABSTRACT

The main focus of this PhD thesis is a part of a research domain of present interest in the materials Science and its biomedical applications. This domain contains the elaboration and study of conducting polymer film based macro-scale and micro-scale level sensors. As a proof of concept, we have conducted a dynamic study on conducting polymer film based bioelectrode for the quantitative detection of human cardiac biomarker, myoglobin (cMb). These bioelectrodes have been used as both impedimetric and chemiresistive sensors.

Herein, we have reported a comprehensive study of the surface morphological and A.C. impedimetric properties of the electrochemically prepared conducting copolymer film poly(pyrrole-co-pyrrolepropylic acid) (PPy-PPa) and its use in device fabrication for biomedical applications. We have shown that copolymer film, PPy-PPa, behaves as a good conducting matrix in both macro and micro-scale sensors. We observe that the impedance response of the PPy-PPa film based bioelectrode is predominantly governed by charge transfer characteristics with major changes in the charge transfer resistance (ΔR_{et}) and very small changes in capacitance, showing strong impedimetric sensitivity towards immunoreaction.

In order to provide mechanical stability and improved charge transport properties of the bioelectrode, we have modified the electrode surface by providing a conducting reduced graphene oxide (RGO) support to the PPy-PPa film. The RGO support provides a conducting and high accessible surface area for the electrochemical deposition of the copolymer. The surface morphology of PPy-PPa-RGO is found to change from an intercalated rope-like structure of PPy-PPa to a porous and inhomogeneous surface, possessing better solution ion diffusion characteristics. It is found that the high protonation level of PPy-PPa contributes to the good ionic and electronic transport ability to the RGO supported conducting matrix. The EIS studies of the PPy-PPa-RGO based bioelectrode show dominant R_{et} characteristics towards target Ag-cMb on immunoreaction in low frequency region of <10 Hz indicating a good biocompatible feature.

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We have further modified the electrode surface by incorporating the metal nanoparticles (MNP) in the conducting polymer film deposited on RGO matrix. The MNP induces a further increase in both the conductivity and inhomogeneity in the polymer film for enhanced diffusion of ions at low A.C. frequency region. The A.C. impedimetric studies shows that the bioelectrode works as a high performance immunosensor in terms of high sensitivity, dynamic range, reproducibility, and specificity for Ag-cMb. Hence, the synergistic effect of RGO and MNP offered enhanced ionic and electronic transport in the nanocomposite film responsible for an enhanced immunoreaction at the electrode / electrolyte interface, providing a better biosensing performance, suggesting its potential applications in the quantitative detection of Ag-cMb for further study, in the actual clinical environment.

Although above method of cMb detection on a macro-scale level is very sensitive and reliable, the biosensor performance has further been improved comparatively by decreasing the inherent complexity, relatively large sample size, multiple steps, and detection time using a miniaturized electrode system in a micro-scale level. Herein, we have utilized 3 μm apart microfabricated 16 pairs of gold electrodes with each having a dimension of (200 x 100 μm) on SiO_2/Si wafer. The 3 μm gap between the source and the drain electrodes was bridged by A.C. electrophoretic alignment of single-walled carbon nanotubes (SWNT) and the said conducting channel was electrochemically modified with conducting PPy-PPa polymer for the biomolecular immobilization of protein antibody, Ab-Mb, for the direct, ultrasensitive, label free, current based detection of cardiac cMb. A systematic field effect (back gate) charge transfer characteristics (FET) studies conducted over the above biosensing system showed an electrostatic gating effect with a significant charge transfer to SWNT. It is observed that high protein loading with strong covalent binding to PPy-PPa/SWNT leads to an increased immunoreaction with the target Ag-cMb, resulted in ultra-high sensitivity to Ag-cMb.

It is concluded that the ease of preparation with cost effective and biocompatible features, make PPy-PPa suitable for practical biomedical applications in both macro and micro scale clinical diagnostics. The high sensitivity, selectivity and good biocompatibility of the PPy-PPa film based bioelectrode / device can provide a platform for the label free detection of the target biomolecules, after appropriate optimization with real blood / serum, for clinical diagnosis in future.

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