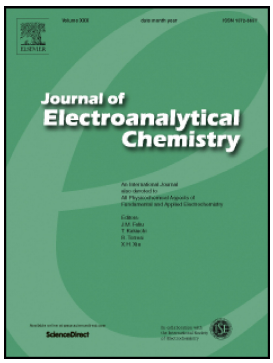

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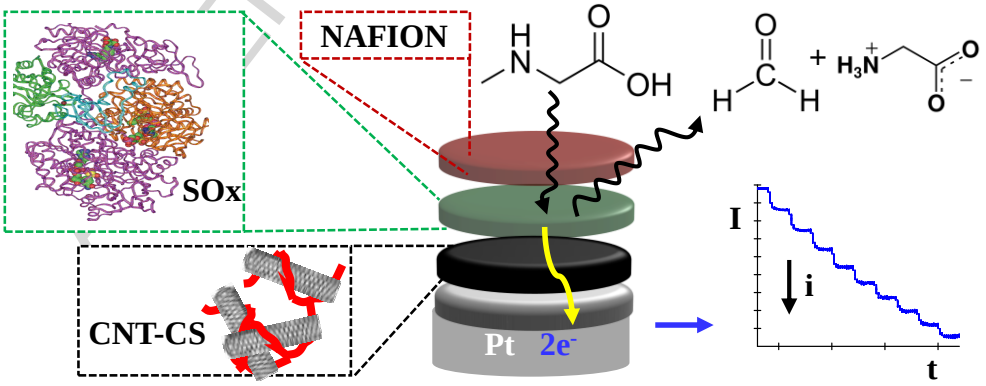
Versatile Sarcosine and Creatinine Biosensing Schemes Utilizing Layer-by-Layer Construction of Carbon Nanotube-Chitosan Composite Films

Michael J. Pannell, Elizabeth E. Doll, Najwa Labban, Mulugeta B. Wayu, Julie A. Pollock,* and Michael C. Leopold*

Abstract

Layer-by-layer composite films of carbon nanotubes (CNTs) within a chitosan matrix with sarcosine oxidase enzyme and capped with Nafion have been developed and optimized as a versatile 1st generation amperometric sarcosine biosensing platform that operates successfully both as an isolated sarcosine sensor as well as a functional component within a creatinine sensor. Accurate measurement of sarcosine in urine and creatinine in blood may help with early diagnosis of diseases such as prostate cancer and renal failure, respectively. In this study, each material within the film is systematically optimized toward sarcosine sensitivity, including a critical evaluation of different CNTs effect on sensing performance. Films featuring carboxylic acid–modified single–walled carbon nanotubes and strategic enzyme doping were shown to be most effective sarcosine sensing platforms, exhibiting excellent sensitivity (~0.5 μA/mM), a linear response (≤ 0.75 mM), fast response time (8 s), low limits of detection (~6 μM), as well as both continuous use stability (7 days) and effective shelf life (> 12 days). Operation of the sarcosine sensor was demonstrated in a sarcosine-spiked urine matrix, detecting the analyte at physiologically relevant concentrations (≥ 20 μM) and successfully quantifying sarcosine-spiked urine samples at 20, 40, and 90 μM (n = 9, 7, and 3) with high average percent recoveries (> 98%) and low relative error. The sarcosine sensing platform was also adapted to a 1st generation creatinine biosensing scheme in which the sarcosine enzymatic reaction is critical to a tri-enzymatic cascade event. The creatinine sensor yielded sensitivity of ~0.6 μA/mM, similar sensing performance parameters to the sarcosine sensor, and was effectively operated in blood serum at physiologically relevant creatinine concentrations (> 1 mM). The demonstrated functionality of these sensors in their respective biological fluids at physiological concentrations of the analyte species suggests potential clinical application as diagnostic tools.



Graphical Abstract

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