Abstract (Eng.)

The present Ph.D. thesis describes fundamental research on molecular imprinting, which was oriented towards devising, fabricating, and testing electrochemical sensors for sensitive and selective determination of gamma-aminobutyric acid (GABA), a biomarker of autism spectrum disorder, and *Escherichia coli* K-12 bacterium. Moreover, procedures were developed to synthesize biocompatible polyacrylamide gel and (inorganic core)-(gel shell) particles in aqueous systems.

An MIP film was deposited on a Pt disk electrode and an Au interdigitated electrode (IDE) array to engineer a chemosensor for GABA biomarker detection and selective determination. The *bis*-bithiophene derivatized functional monomers were chosen to devise an MIP for GABA. The stability of the pre-polymerization complexes of the GABA template with *bis*-bithiophene derivatized functional monomers was estimated with the density functional theory (DFT) at the B3LYP level. Different electrochemical transductions, including differential pulse voltammetry (DPV), capacitive impedimetry (CI), and electrochemical impedance spectroscopy (EIS), were tested to evaluate the MIP chemosensor performance. XPS measurements confirmed the GABA template complete extraction from the MIP film. The morphology of the MIP film deposited on IDEs was unraveled and characterized by AFM. For DPV, CI, and EIS electrochemical transductions, the sensitivity, selectivity, and limit of detection (LOD) of the MIP chemosensors were calculated. Furthermore, GABA in artificial serum samples was determined to confirm the chemosensor suitability for clinical analysis. The EIS transduction appeared most suitable for potential point-of-care GABA determinations.

A modified Gram-negative strain of *Escherichia coli* K-12, i.e., E2152, was determined using the herein devised capacitive impedimetric MIP chemosensor. The extracellular matrix (ECM) of E2152 strongly interacted with a boronic acid group because of the porous and flexible polymers of the cell wall. The scanning electron microscopy (SEM) and atomic force microscopy (AFM) imaging confirmed E2152 template entrapment in the MIP and then the effectiveness of the template extraction. Moreover, the X-ray photoelectron spectroscopy (XPS) measurements, as well as DPV and EIS transductions, confirmed the E2152 template extraction. The E2152 analyte binding was then demonstrated using CI. The interference study, performed using *E. coli* variants expressing different surface appendages (type 1 *fimbriae* or Antigen 43 protein) or *Shewanella oneidensis* MR-1, another Gram-negative bacterium, demonstrated that the bacterial surface composition notably impacts sensing properties of the E2152-templated MIP chemosensor.

A one-step electrochemically initiated gelation was used to synthesize biocompatible polyacrylamide gel microparticles and core-shell nanoparticles. In this gelation, electrochemical decomposition of ammonium persulphate initiated copolymerization of the *N*-isopropylacrylamide, methacrylic acid, and *N,N'*-methylenebisacrylamide monomers under hydrodynamic conditions. SEM and TEM imaging confirmed the fabrication of gel particles. The particles were thoroughly characterized with NMR spectroscopy, TGA, BET, and DLS measurements. They were explored to prepare 3D cell-like structures.

The lyophilized polyacrylamide gel microparticles and core-shell nanoparticles were applied to form a 3D culture of MDA-MB-231 and HeLa cells using gel embedment. The nanoparticles' biocompatibility and IC_{50} values were confirmed using MTT assays. The confocal microscopy showed that core-shell nanoparticles provided superior support for complex 3D tissue structures.