Project Summary

Overview

Tetrahedral amorphous carbon (t*a*-C) and nitrogen-incorporated tetrahedral amorphous carbon (t*a*-C:N) thin-films have emerged as a next-generation electrode that could prove useful for a variety of electroanalytical applications. t*a*-C and t*a*-C:N materials are a diamond-like carbon that consist of a mixture of sp^2 and sp^3-bonded carbon. They exhibit many of the excellent properties (wide potential window, low background current, good responsiveness without conventional pretreatment and microstructural stability) of boron-doped diamond but can be deposited (200-400 nm thickness) by physical vapor deposition near room temperature. Deposition times are tens of minutes. This contrasts with the harsher chemical vapor deposition conditions required for boron-doped diamond including hours of growth time and temperatures in the 700-800 ^{o}C range. In electroanalysis, specifically electrochemical detection coupled with flow injection analysis (FIA), t*a*-C:N electrodes offer significant improvements in the analytical detection figures of merit (linear dynamic range, limit of detection, response reproducibility and response stability) for analytes as compared to conventional sp^2 carbon electrodes (*e.g.*, glassy carbon). These electrodes are useful for detecting chemical contaminants in water supplies, particularly analytes that require high positive potentials for oxidation. The electrodes also resistant to fouling. So far, there has been little research reported on the basic electrochemical properties of these electrodes in aqueous electrolytes and none in organic electrolyte/solvent systems. To this end, research in this project will generate new science on how capacitance, molecular adsorption and electron-transfer kinetics in aqueous and organic electrolytes correlate with the sp^2/sp^3 carbon ratio and nitrogen content of the films. New electroanalytical applications using flow injection analysis and reversed-phase LC-EC will be explored for target contaminants in water supplies that are challenging to detect with conventional sp^2 carbon electrodes. The research will be focused on three specific aims.

Intellectual Merit

The planned fundamental research will advance knowledge about these new carbon electrodes regarding the structure of electrified interfaces and heterogeneous electron-transfer kinetics for soluble and surface-confined redox systems in aqueous and organic electrolytes. Understanding material property-electrochemical behavior relationships is critical for developing new electroanalytical applications for t*a*-C and t*a*-C:N thin-film electrodes that benefit the environment and human health. The effort will produce new insights on how the material properties, diffusion and electrochemical reaction rate impact the electrode performance. Heterogeneous electron-transfer rate constants and activation energies for electron-transfer for various redox probe molecules will be determined in different aqueous and organic solvent/electrolyte systems. All work represents new science that will favorably impact the field of electroanalytical chemistry.

Broader Impacts

Michigan State University has a long history of excellence in integrating its world-grant mission to apply teaching, research and outreach toward improving the social and environmental condition of humanity. The proposed research will produce knowledge about the physical, chemical and electrical properties of a new carbon electrode - *tetrahedral amorphous carbon*. These electrodes are potentially useful for a range of electrochemical applications beneficial to human health and the environment. Knowledge and practices gained from the project's interdisciplinary research agenda will give AAP participant the tools needed to be a successful scientist and academician. The AAP Fellow will develop improved critical thinking and communication skills and learn about productive teamwork in a laboratory setting. Planned professional development activities will give the Fellow confidence and make them more competitive for employment. The AAP Fellow will experience the benefits of positive mentoring and understand how proper mentor-mentee relationships should exist. The Fellow will be educated on laboratory safety and proper chemical hygiene practices. Finally, the Fellow will work with the PI to develop lectures and exams and to deliver lectures in an undergraduate course on *Instrumental Methods of Analysis* (SS2021, NEU 417).