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Neutral Red Redox Polymer/graphene quantum dots composite prepared in natural deep eutectic solvents for sulfanilamide sensing

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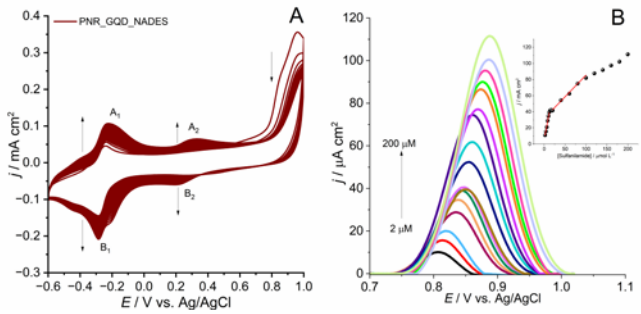
Poly neutral red (PNR) is a redox polymer typically prepared by electropolymerization of monomers, initiated by a radical cation via C-N coupling [1]. Electrochemical sensors based on these polymers have been studied due to their rapid electron transfer and electrocatalytic properties, which ensure more sensitive and selective methods [1 -2] as the electrochemical properties of the polymeric film can be controlled by the composition of the electropolymerization medium. The use of natural deep eutectic solvents (NADES) was as an alternative to ionic liquids and traditional solvents, improving the stability and uniformity of the film in an environmentally friendly manner [2-3]. These redox polymers can also be combined with nanomaterials to form nanocomposites. In this context, the graphene quantum dots (GQDs), have increased attention for developing new nanocomposites in electrochemistry due to their easy synthesis, environmental friendliness, and high electroactive surface area [4]. The combination of redox polymers, GQDs, and NADES in electrochemical applications remains unexplored, presenting a promising future due to the enhanced electrochemical response, higher detection limits, and sensitivity to emerging pollutants such as sulfonamides can be found in water sources and risk to ecosystems and human health. This work presents a novel electrochemical sensor based on PNR-GQD in NADES for the detection of sulfonamides. The modified sensor was characterized using cyclic voltammetry (CV), impedance spectroscopy, and scanning electron microscopy. The GQDs were characterized by transmission electron microscopy, UV-Vis spectroscopy, and Fourier-transform infrared spectroscopy. The NADES were prepared with hydrogen bond acceptors (Choline Chloride) and hydrogen bond donors (fructose, glucose, acetic acid, or citric acid) in a 2:1 molar ratio, and GQD by citric acid pyrolysis. The GQD dispersed in NADES presented a diameter of 3 nm ± 0.7. In all NR CVs obtained in NADES, two well-defined redox couples appear, where the pair (A_2/B_2) that can be attributed to the oxidation/reduction of the polymer and monomer presents a current increase with the increase in the number of cycles. The best polymer films were obtained in NADES based on acetic acid with 0.1 M ammonium acetate and 2 mg mL^{-1} of GQD in Fig 1a. The NADES and GQD enhance their electrochemical response through complementary electrical, electrochemical, and mechanical properties, resulting in a more uniform polymeric film observed in FEG-SEM analysis. The nanocomposite based on the polymeric film led to increased electrochemical performance for the detection of sulfanilamide with a low LOD (0.29 µmol L^{-1}) and a linear range (2-140 µmol L^{-1}) in Fig 1b. This demonstrates that poly (NR)/GQD/NADES nanocomposite film-modified electrodes are very promising for future electrochemical applications.

Fig 1. a) Potential cycling electrodeposition of PNR films on GCE at a scan rate of 25 mV s^{-1} containing 1.0 mmol L^{-1} NR in NADES and GQD. b) DPVs obtained with PNR-GQD-NADES in ammonium buffer pH 9.0 for 2.0 to 200 µmol L^{-1} and inset corresponding calibration plot.



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