Electrogeneration of carbon-based nanomaterials with tunable redox properties for nanomedicine

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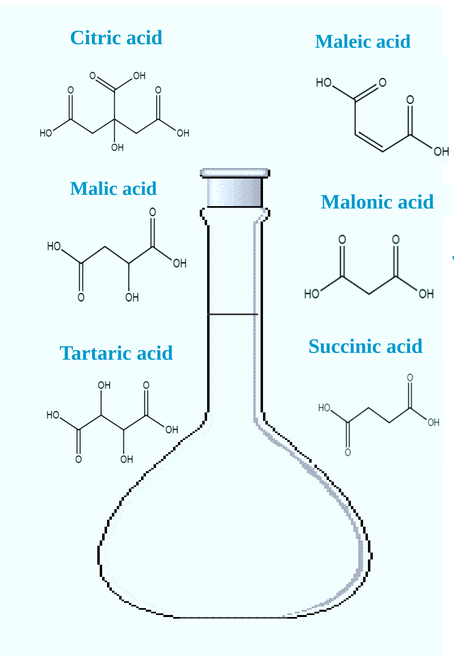
Introduction and Aims

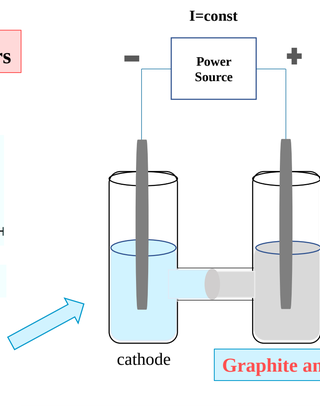
The application of nanomaterials in medicine comprises its use for diagnosis, monitoring, control, prevention, and treatment of diseases. In the therapeutic applications, nanomaterials are essentially applied as drug vehicles, however, they can also be applied as the drug itself by taking advantage of their intrinsic properties as water-solubility, capacity to diffuse in the intracellular aqueous medium and redox-properties, i.e., ability to work as electron donors/ electron-acceptors in an appropriate potential range [1]. Previous studies with water-soluble carbon-based nanomaterials produced electrochemically from graphite (named Electrogenerated Hydrophilic Carbon-EHC) revealed an electrochemical behavior that is strongly dependent on the buffer electrolyte used in the preparation of the EHC nanomaterial, i.e., in a phosphate buffer solution, the generated nanomaterial exhibits electron-accepting capacity but when a citrate buffer is used it shows electron-donor ability [2,3]. This work aims to understand the effect of the structure of the carboxylic acid buffer (biocompatible) used in the EHC generation on its redox properties and evaluate if it is appropriate for mimicking the enzymes involved in cell redox balance and signaling.

Experimental Design

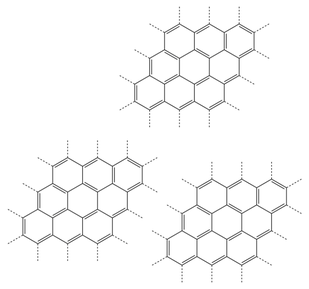
Electrolytes: 6 different Carboxylic acid buffers



Ionic strength: 0.40; pH: 4.5-6.5



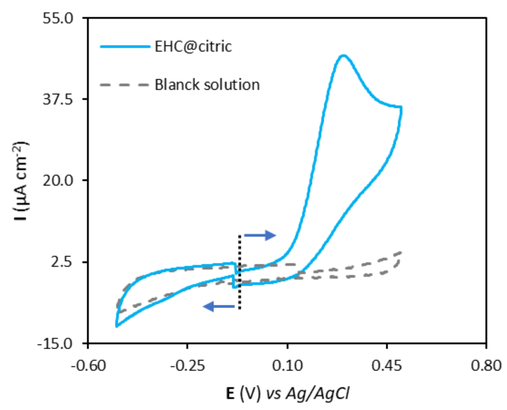
EHC nanomaterials are electrochemically generated from a graphite anode in the different carboxylic buffer electrolytes



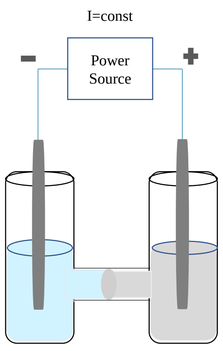
EHC nanomaterials solution

Cyclic voltammetry characterization

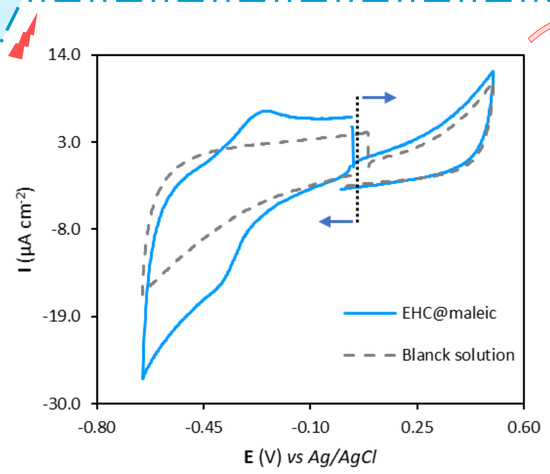
Results



Citric acid

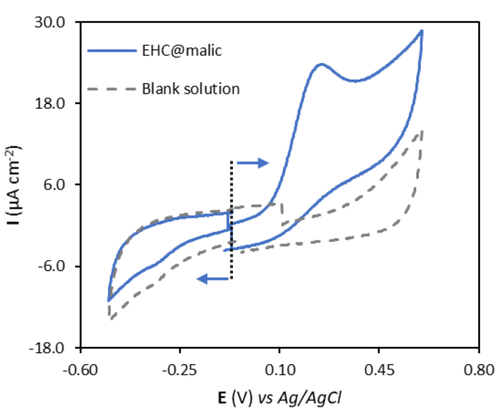


Maleic acid

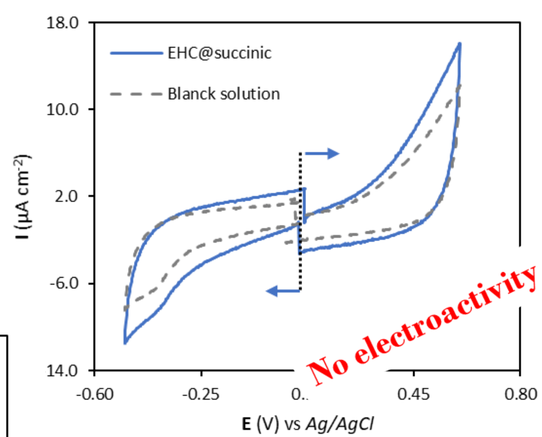


EHC generated in an oxidized state with electron-accepting properties

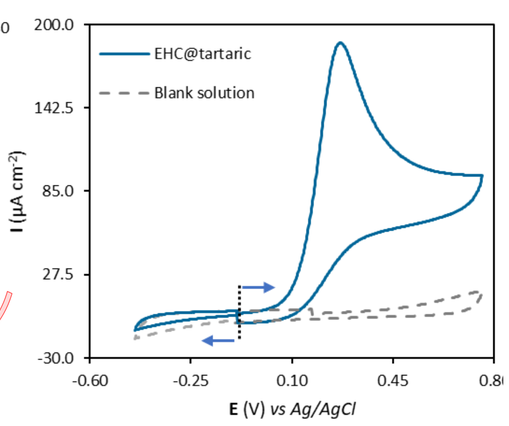
Malic Acid



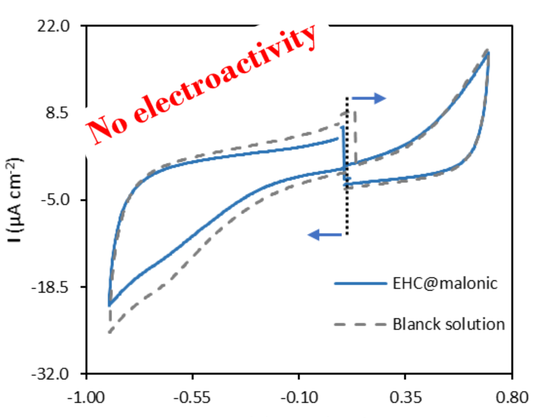
Succinic acid



Tartaric acid

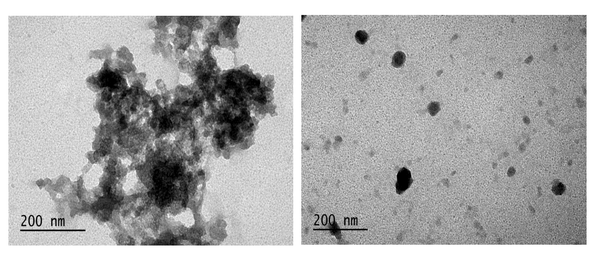


Malonic acid



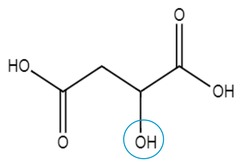
EHC generated in a reduced state with electron-donating properties

TEM analysis



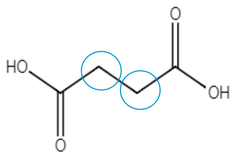
Conclusions

Dicarboxylic acids with α-C(OH) groups



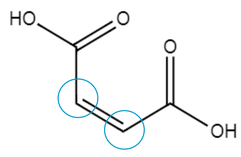
Irreversible electron-donating properties

Dicarboxylic acids with α-CH_2- groups



No electroactivity

Dicarboxylic acid with α-CH- group



Quasi-reversible electron-accepting properties

EHC operate in a potential range (0 to 0.7 V) compatible with the different substrates of enzymes involved in cell redox balance and signaling, e.g., SOD= 0.42 V (*vs Ag/AgCl*).

Future work

- ✓ Evaluate the EHC nanomaterial's ability to mimic, in aqueous buffers, the activity of enzymes involved in cell redox balance and signaling (e.g., Peroxidases, Superoxide Dismutase, Catalase, Glutathione reductase), and characterize, their catalytic mechanisms and kinetics;
- ✓ Assess the cytotoxicity of EHC nanomaterials and evaluate their ability to restore the redox state of cells under oxidative stress conditions.

References

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