Amino acid-based hydrogels for electrochemical applications

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INTRODUCTION

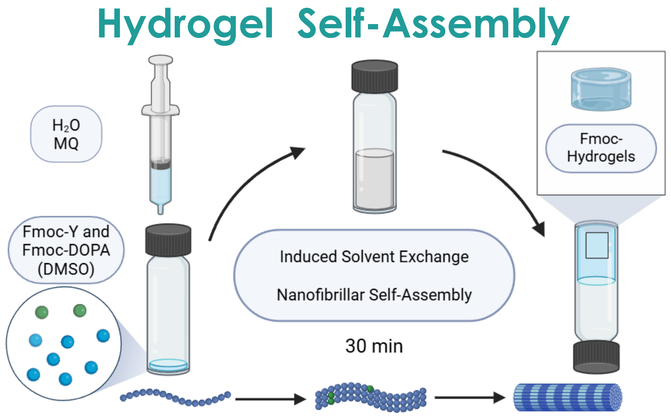
Soft bioelectronics require electrolyte materials that are biocompatible, flexible, ionically conductive, electrically isolating, and electrochemically stable. Conventional liquid electrolytes can suffer from leakage and poor mechanical integration, motivating the development of solid-state alternatives [1].

Fmoc-protected amino acids are promising low-molecular-weight building blocks because they self-assemble into hydrated supramolecular networks through aromatic π–π stacking and side-chain interactions [2,3]. Compared with larger protein-based biomaterials, these systems offer simpler molecular design, easier processing, and tunable functionality [4].

In this work, Fmoc-Tyrosine (Fmoc-Y) and Fmoc-Tyrosine:DOPA (Fmoc-YD) hydrogels were explored as supramolecular amino acid-based electrolyte matrices for soft bioelectronic applications. By comparing their self-assembly behavior, structural features, and electrochemical performance, this study aims to assess whether these low-molecular-weight hydrogels can function as simple and tunable solid-state electrolyte candidates.

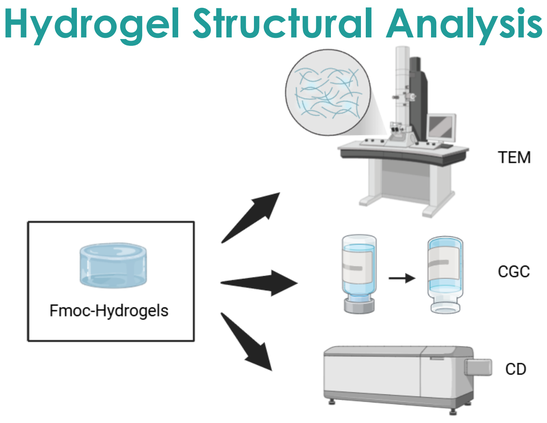
MATERIALS AND METHODS

Hydrogel Self-Assembly



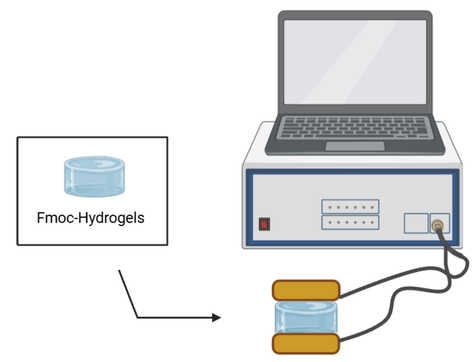
Fmoc-Y and Fmoc-YD (3:1 ratio) ranging from 2 to 10 mg/mL. Solvent exchange-triggered self-assembly using MiliQ water. Samples left to stand for 30 min to gelate.

Hydrogel Structural Analysis



Fmoc-hydrogels' structural analysis comprised Transmission Electron Microscopy (TEM), Circular Dichroism (CD) and Critical Gelation Concentration (CGC) assays.

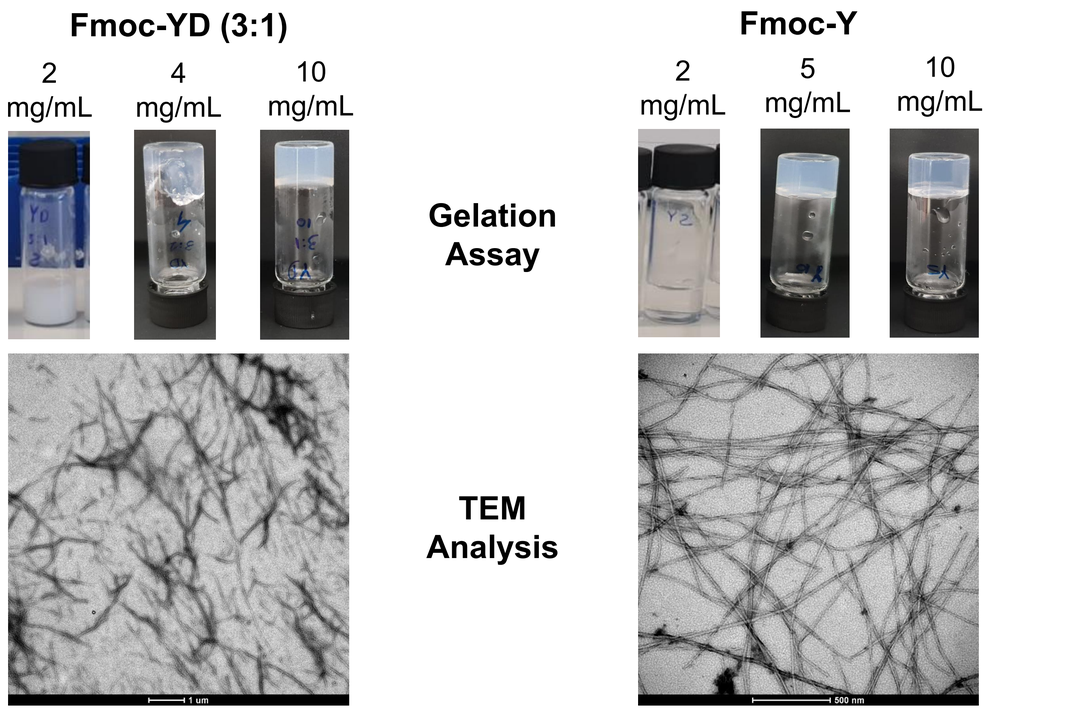
Solid-state Electrochemical Analysis



Solid-state electrochemical analysis was performed using a two-electrode (Gold) sandwich setup inside a Faraday cage. Open circuit potential (OCP), Impedance spectroscopy (EIS) and Voltammetry assays were conducted (CV; LSV).

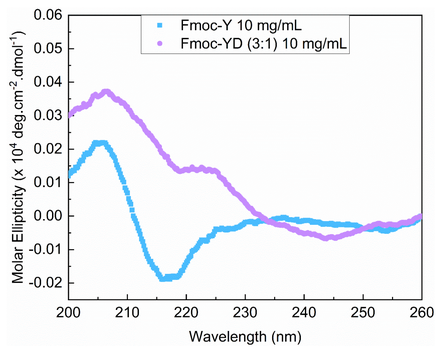
RESULTS

Fmoc Hydrogels Structural Characterization



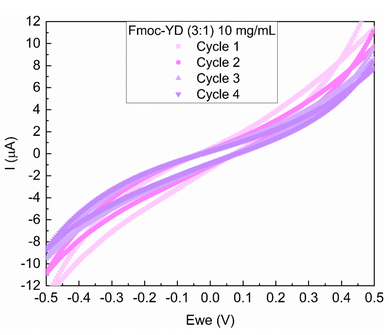
Dispersed fiber network (10 mg/mL)

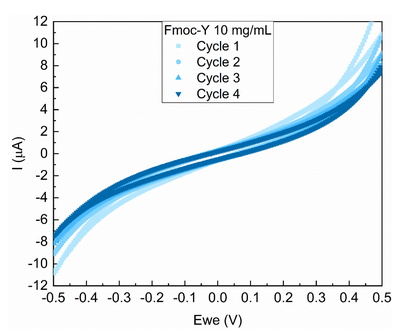
Densely packed fiber network (10 mg/mL)

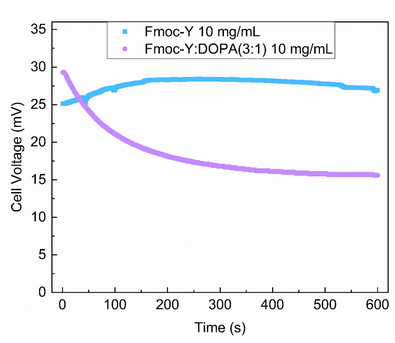


Evidence of β-sheet supramolecular configuration, more pronounced in Fmoc-Y with a minimum at ~215 nm.

Fmoc Hydrogels Solid-state Electrochemical Analysis

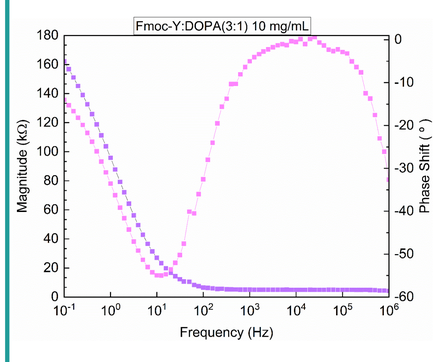


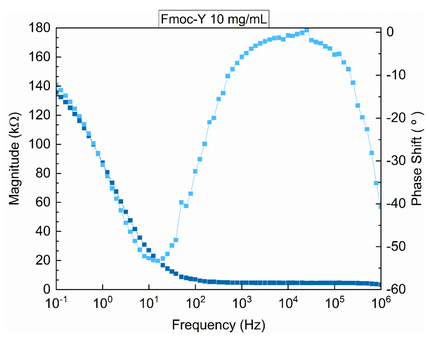




Absence of distinct redox peaks suggests ion-mediated interfacial response

Both materials reach a stable electrochemical response over time





- Ion-conducting behavior
- Phase shift → Bulk ionic conduction to electrode polarization
- High Hz plateau → Bulk resistance
- Low Hz → Electrode Polarization

System	Method	Source of conductivity (σ)	σ (S/cm)
Fmoc-Y 10 mg/mL	LSV (DC)	Apparent	4.5E-06
	EIS (AC)	Bulk	9.0E-05
Fmoc-YD (3:1) 10 mg/mL	LSV (DC)	Apparent	4.9E-06
	EIS (AC)	Bulk	8.3E-05

Both hydrogels show measurable ionic conductivity; EIS provides bulk conductivity, while LSV reflects apparent DC conductivity influenced by polarization, supporting their potential as soft electrolyte materials.

MAIN CONCLUSIONS AND FUTURE WORK

- ✓ Fmoc-hydrogels form fibrillar networks with β-sheet supramolecular organization and stable ion-conducting behavior with conductivities of 10^{-5} S/cm.
- ✓ Fmoc-Y appears as the more promising system, with higher conductivity and structural stability.
- ✓ Fmoc-hydrogels appear as electrolyte-like ion-conducting matrices promising for low-current electrochemical applications.
- Future work will include the structural and electrochemical evaluation of other Fmoc-protected amino acids.

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