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CMC-AC-CB-Based Separator-Free Quasi-Solid Supercapacitors: Bifunctional Polymer Architecture Enabling Fluorine-Free Grid-Scale Energy Storage for Decarbonization

Thiago Ferro de Oliveira[1–4]* and Simoni Margareti Plentz Meneghetti[2–3]

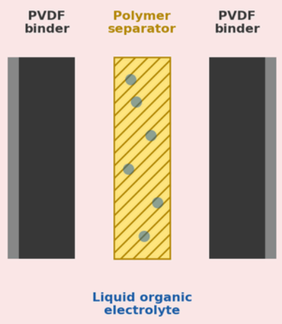
[1] Institute of Mathematics [2] Postgraduate Program in Chemical Engineering [3] GCAR – Chemistry Department [4] Postgraduate Program in Renewable Energies

*Federal University of Alagoas (UFAL), Maceió / Rio Largo — AL, Brazil · * thiago.oliveira@im.ufal.br*

INTRODUCTION & AIM

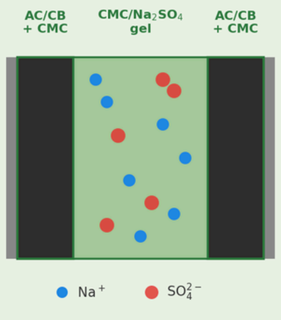
Bifunctional CMC: A Single Bio-Polymer for the Whole Device

CONVENTIONAL APPROACH



- ✗ Fluorinated PVDF binder
- ✗ Toxic NMP solvent
- ✗ Synthetic separator
- ✗ Multiple incompatible materials

THIS WORK — CMC DUAL ROLE



- ✓ CMC binder (bio-derived)
- ✓ Aqueous green processing
- ✓ Separator-free architecture
- ✓ CMC matrix = chemical compatibility

AIM

Develop a fully bio-derived, fluorine-free, separator-free quasi-solid supercapacitor by exploiting carboxymethylcellulose (CMC) as a single bifunctional polymer — simultaneously the electrode binder and the gel-electrolyte matrix — assembled via a sequential layer-by-layer route compatible with grid-scale energy storage.

METHOD

Layer-by-Layer Quasi-Solid Cell Assembly

1. AQUEOUS GREEN SLURRY

Activated Carbon (AC) + Carbon Black (CB) + CMC (binder, water-based)

Mass ratio 80 : 10 : 10 • Solvent: H_2O (no NMP)

2. ULTRASONIC HOMOGENIZATION

Cavitation breaks agglomerates and disperses amphiphilic CMC over carbon particles

Sonication 10 – 30 min • Forms percolation pathways

3. CASTING ON CURRENT COLLECTOR

Slurry doctor-bladed onto metal foil → first electrode layer formed in situ

Direct casting • No transfer step

4. CONTROLLED PRE-DRYING

Removes free water • preserves coordinated water (critical for Na^+ solvation & ionic mobility)

T = 60 – 70 °C • Free-water peak ≈ 62.76 °C

5. CMC / Na_2SO_4 GEL DEPOSITION

Viscous polyelectrolyte gel poured directly on the pre-dried electrode → integrated interface

Acts as electrolyte AND separator (no membrane!)

6. SECOND ELECTRODE + PRESSING

Symmetric counter-electrode applied on top; sandwich consolidated under contact pressure

P = 0.5 MPa • conformal contact, ↓ ESR

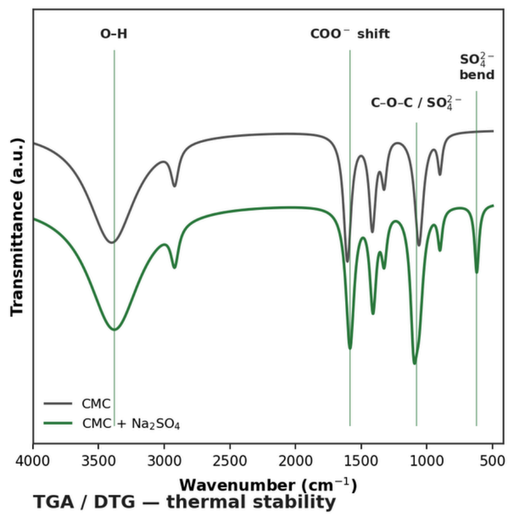
→ ASSEMBLED, SEPARATOR-FREE QUASI-SOLID-STATE SUPERCAPACITOR ←

RESULTS & DISCUSSION

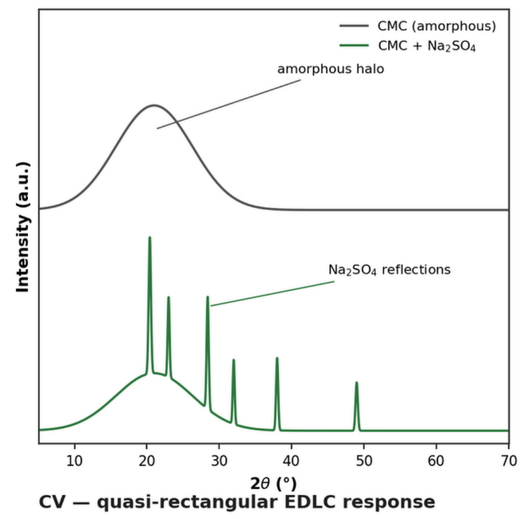
EXPECTED CHARACTERIZATION SIGNATURES — predicted from literature precedent

Profiles below illustrate the diagnostic signatures the protocol is designed to produce, informed by Refs. [1, 2, 6, 7, 8].

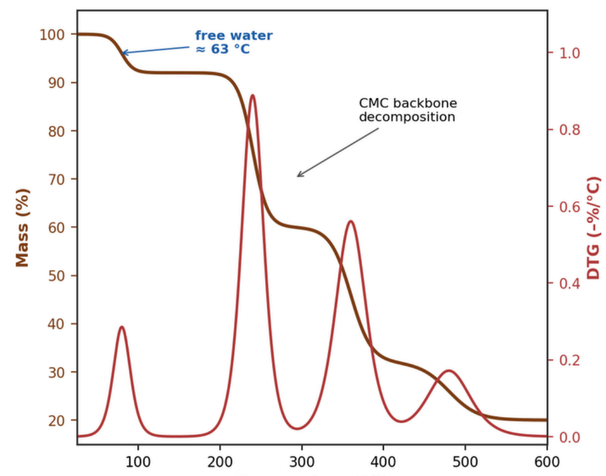
(a) FTIR — polymer–salt complexation



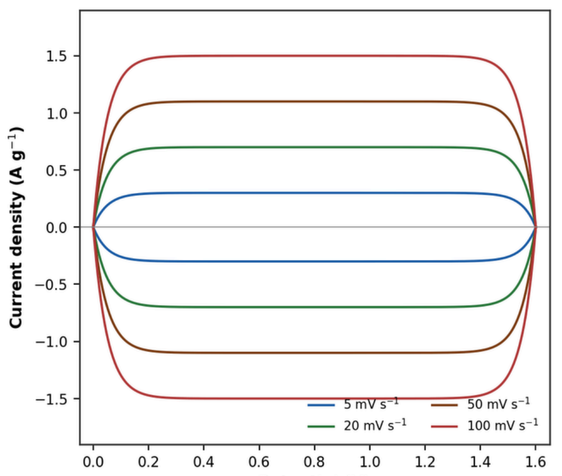
(b) XRD — amorphicity + ionic loading



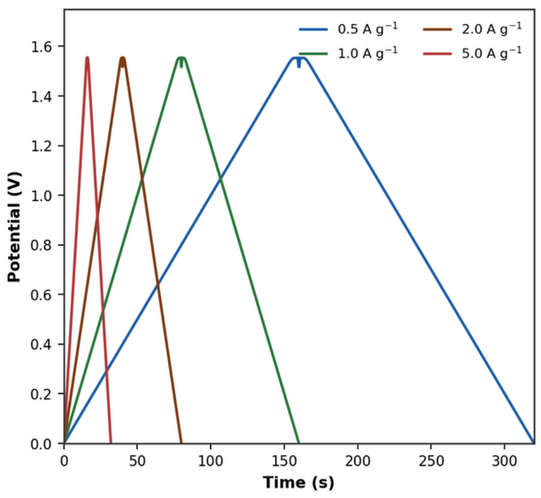
(c) TGA / DTG — thermal stability



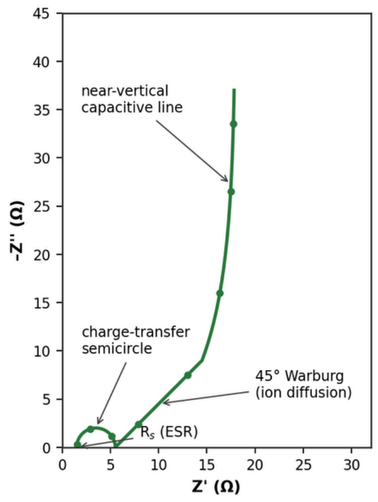
(d) CV — quasi-rectangular EDLC response



(e) GCD — galvanostatic charge/discharge



(f) EIS — Nyquist (100 mHz – 10 kHz)



Interpretation criteria for the proposed protocol:
- FTIR shift of COO^- asymmetric band ⇒ Na^+-CMC complexation. • Broad XRD halo ⇒ amorphous, ion-permeable matrix.
- Resolved free-water peak (≈63 °C) AND retained mass to ≈200 °C ⇒ coordinated-water preserved.
- Quasi-rectangular CV + triangular GCD + small R_s and 45° Warburg ⇒ efficient EDLC behavior across the QSS interface.

CONCLUSION

A SINGLE BIO-POLYMER (CMC) ENABLES A FULLY GREEN SUPERCAPACITOR ARCHITECTURE

Fluorine-FREE	BIO-DERIVED	SEPARATOR-FREE	DUAL ROLE	GRID-SCALE READY
no PVDF / NMP solvents	CMC binder + gel matrix	gel acts as membrane	single polymer throughout cell	aqueous, scalable layer-by-layer

FUTURE WORK

- Experimental validation of the proposed protocol across AC/CB:CMC ratios and Na_2SO_4 molarities
- Long-term cycling stability and self-discharge benchmarks at full pouch-cell scale
- Optimization of pre-drying window to maximize coordinated-water retention vs. mechanical strength
- Extension to alternative neutral aqueous salts (e.g. Li_2SO_4, K_2SO_4) and bio-derived dopants

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