

## Supplementary Materials

# Sustainable and printable nanocellulose-based ionogels as gel polymer electrolytes for supercapacitors

Rosa M. González-Gil <sup>1,2,†</sup>, Mateu Borràs <sup>3,4,†</sup>, Aiman Chbani <sup>3</sup>, Tiffany Abitbol <sup>5</sup>, Andreas Fall <sup>5</sup>, Christian Aulin <sup>5,6</sup>, Christophe Aucher <sup>3,\*</sup> and Sandra Martínez-Crespiera <sup>1,\*</sup>

<sup>1</sup> Applied Chemistry and Materials, ARTS Department, Leitat Technological Center, C/Pallars, 186-179,

08005 Barcelona, Spain; rosamaria.gonzalez@icn2.cat

<sup>2</sup> Novel Energy-Oriented Materials Group, Catalan Institute of Nanoscience and Nanotechnology, ICN2 (CSIC-BIST), Edifici ICN2, Campus UAB, 08193 Barcelona, Spain

<sup>3</sup> Energy and Engineering, ARTS Department, Leitat Technological Center, C/de la Innovació, 2,

08225, Barcelona, Spain; mateu@bioo.tech (M.B.); achbani@leitat.org (A.C.)

<sup>4</sup> Arkyne Technologies SL (Bioo), C/de la Tecnologia, 17, 08840, Barcelona, Spain

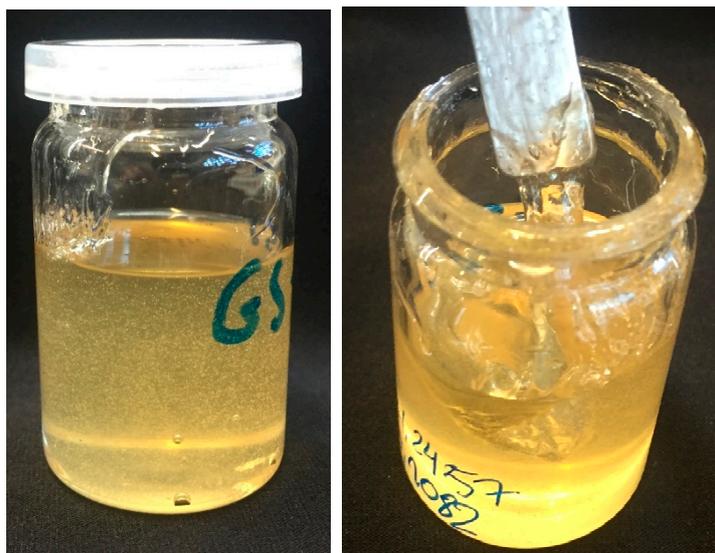
<sup>5</sup> Bioeconomy and Health, RISE Research Institutes of Sweden, Drottning Kristinas väg 61,

114 28 Stock-holm, Sweden; tiffany.abitbol@ri.se (T.A.); [andreas.fall@ri.se](mailto:andreas.fall@ri.se) (A.F.); [Christian.aulin@Holmen.com](mailto:Christian.aulin@Holmen.com) (C.A.)

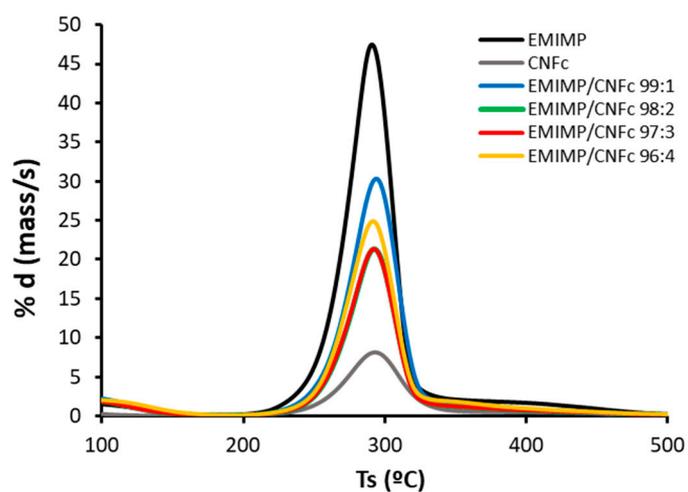
<sup>6</sup> Holmen Iggesund, 825 80 Iggesund, Sweden

\* Correspondence: [caucher@leitat.org](mailto:caucher@leitat.org) (C.A.); [sandramartinez@leitat.org](mailto:sandramartinez@leitat.org) (S.M.-C.); Tel.: +34 93 788 23 00 (S.M.-C.)

† These authors contributed equally to this work.



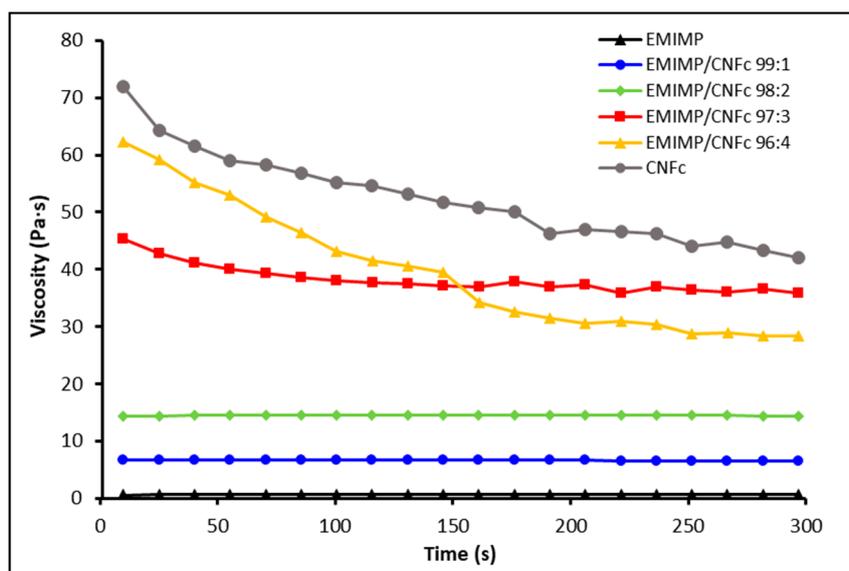
**Figure S1:** Final aspect of the as-prepared ionogels pictures.



**Figure S2:** Differential scanning calorimetry thermogram (DSC) of the EMIMP/CNFc mixtures (99:1, 98:2, 97:3 and 96:4). Pristine CNFc and EMIMP were added for comparison purposes.

|                 | EMIMP/NC ratio | T decomposition (°C) |
|-----------------|----------------|----------------------|
| EMIMP           | 100:0          | 290,0                |
| CNFc            | 0:100          | 294,2                |
| EMIMP/CNFc 99:1 | 99:1           | 292,0                |
| EMIMP/CNFc 98:2 | 98:2           | 292,3                |
| EMIMP/CNFc 97:3 | 97:3           | 292,7                |
| EMIMP/CNFc 96:4 | 96:4           | 293,0                |

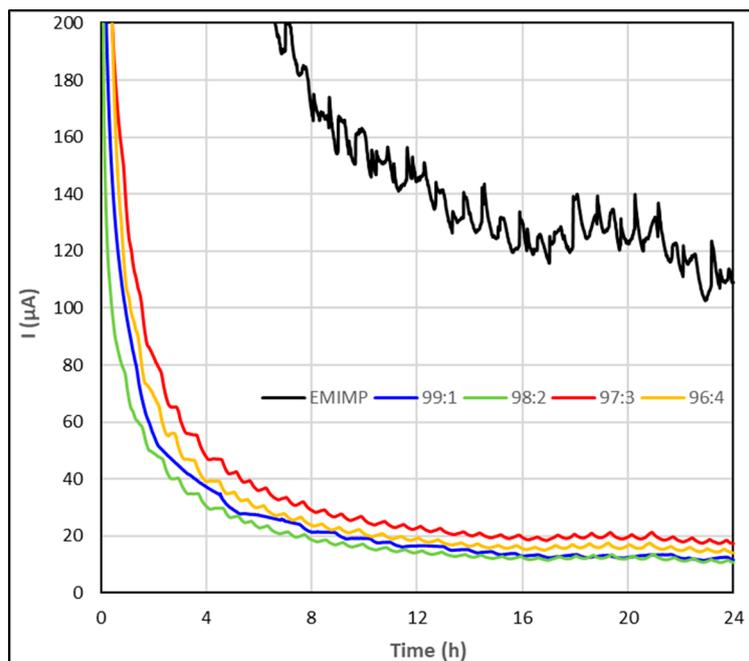
**Table S1:** Temperature decomposition for each EMIMP/CNFc mixtures (99:1, 98:2, 97:3 and 96:4) mixtures, obtained from differential scanning calorimetry thermograms (DSC). Pristine CNFc and EMIMP temperatures are also added.



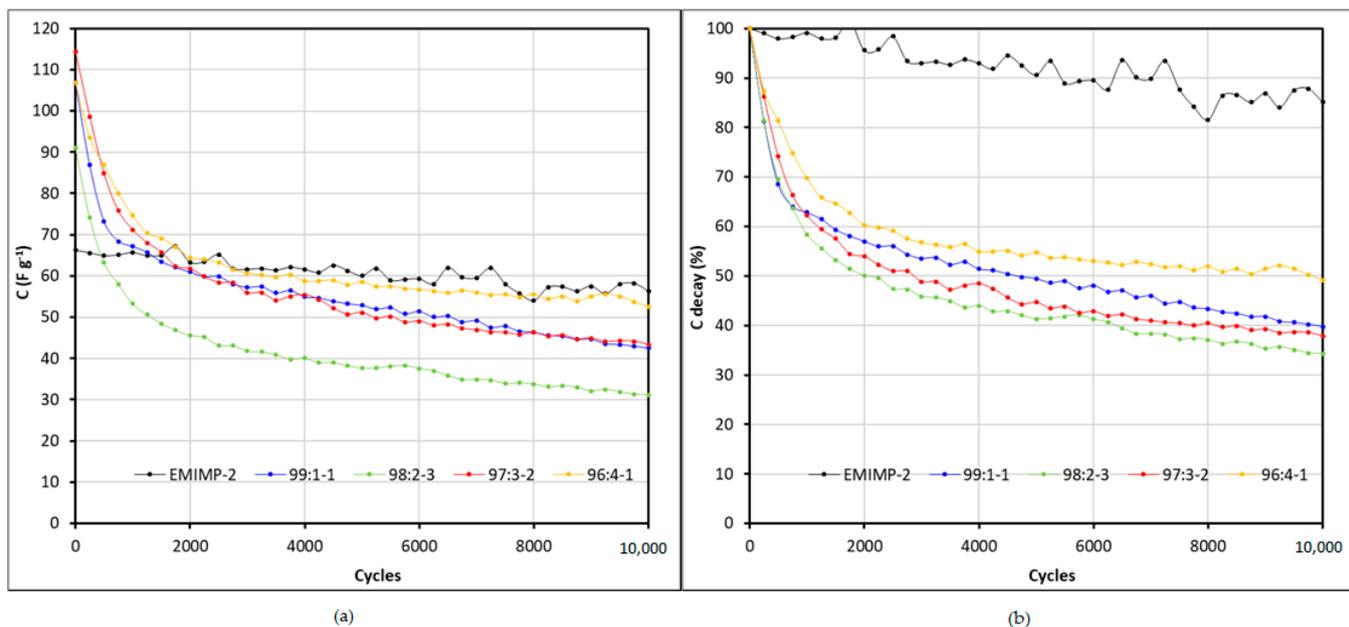
**Figure S3:** Rheological results at room temperature vs. time of the EMIMP/CNFc mixtures (99:1, 98:2, 97:3 and 96:4). Pristine CNFc and EMIMP are added for comparison purposes.

| Electrolyte | $i$<br>(mA/cm <sup>2</sup> ) | $C_e$ (F<br>g <sup>-1</sup> ) | ESR ( $\Omega$<br>cm <sup>-2</sup> ) | $E_{max}$<br>(Wh kg <sup>-1</sup> ) | $P_{max}$ (W kg <sup>-1</sup> ) |
|-------------|------------------------------|-------------------------------|--------------------------------------|-------------------------------------|---------------------------------|
| EMIMP       | 0.5                          | 109±3                         | 15±1                                 | 15.2±0.5                            | 5872.2±490.3                    |
|             | 1                            | 99± 4                         | 14.7±<br>0.7                         | 13.5±0.7                            | 5924.9±485.5                    |
|             | 2                            | 75±4                          | 14.6±0.7                             | 10.2±0.7                            | 6003.3±475.9                    |
|             | 5                            | 35±4                          | 14.7±0.5                             | 5.5±1.4                             | 5974.7±473.5                    |
|             | 10                           | 10±3                          | 15.4±0.6                             | 1.5±0.9                             | 5908.8±764.1                    |
| EMIMP99:1   | 0.5                          | 159±7                         | 11±2                                 | 22.7±1.2                            | 9536.2±2219.8                   |
|             | 1                            | 132± 4                        | 8.2± 0.8                             | 18.7±0.7                            | 11,280.3±2169.9                 |
|             | 2                            | 105±3                         | 7.7±0.8                              | 14.4±0.5                            | 11,796.8±2093.7                 |
|             | 5                            | 59±6                          | 8.4±0.6                              | 7.9±1.1                             | 11,155.5±2209.7                 |
|             | 10                           | 29±7                          | 7.9±0.8                              | 3.6±1.1                             | 10,934±1987                     |
| EMIMP98:2   | 0.5                          | 174±9                         | 11±4                                 | 24.1±1.6                            | 7618.5±2107.4                   |
|             | 1                            | 156± 11                       | 10± 2                                | 21.5±1.8                            | 8132.8±2364.1                   |
|             | 2                            | 127±15                        | 9±2                                  | 17.8±2.5                            | 8311.8±2479.6                   |
|             | 5                            | 73±21                         | 10±2                                 | 10.3±3.6                            | 8107.3±2487.9                   |
|             | 10                           | 30±16                         | 10±2                                 | 4.2±2.7                             | 7695±2462                       |
| EMIMP97:3   | 0.5                          | 172±6                         | 12±1                                 | 24.0±1.2                            | 7677.7±938                      |
|             | 1                            | 160± 6                        | 10.2±<br>0.7                         | 22.3±1.2                            | 8841.9±790.6                    |
|             | 2                            | 139±9                         | 9.7±0.5                              | 19.3±1.8                            | 9330±659                        |
|             | 5                            | 93±12                         | 9.6±0.3                              | 12.9±2.4                            | 9332.7±350.2                    |
|             | 10                           | 44±9                          | 10.2±0.2                             | 6.2±1.8                             | 8823±525                        |
| EMIMP96:4   | 0.5                          | 165±4                         | 7.4±0.5                              | 22.5±0.8                            | 12,717±986                      |
|             | 1                            | 152± 5                        | 5.6± 0.6                             | 20.8±0.9                            | 16,023±1524                     |
|             | 2                            | 130±8                         | 5.3±0.3                              | 18.1±1.3                            | 16,729.1±1723.5                 |
|             | 5                            | 91±12                         | 5.4±0.2                              | 13.3±2.0                            | 16,429.1±1767.6                 |
|             | 10                           | 56±11                         | 5.7±0.3                              | 8.5±1.8                             | 15,463.4±1415.7                 |

**Table S2:** Summary of the key performance parameters on different current density such as specific capacitance, ESR,  $E_{max}$  and  $P_{max}$



**Figure S4:** Current ( $\mu\text{A}$ ) vs. time for supercapacitors with EMIMP and EMIMP/CNFc (99:1, 98:2, 97:3 and 96:4) as electrolytes for a period of 24h



**Figure S5:** The capacity variation with the cycles and for supercapacitors with EMIMP and EMIMP/CNFc (99:1, 98:2, 97:3 and 96:4) as electrolytes.

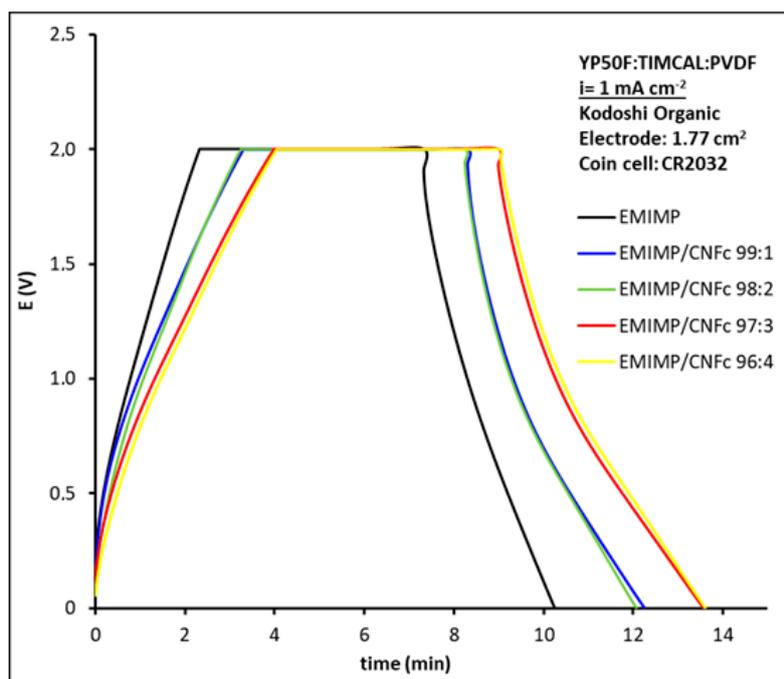
| Electrolyte | $i$ (mA/cm <sup>2</sup> ) | $C_e$ (F g <sup>-1</sup> ) | ESR ( $\Omega$ cm <sup>-2</sup> ) |
|-------------|---------------------------|----------------------------|-----------------------------------|
| EMIMP/CMC   | 0.5                       | 153±3                      | 11.8±1                            |
|             | 1                         | 123± 4                     | 10.6± 0.7                         |
|             | 2                         | 96±4                       | 10.3±0.7                          |
|             | 5                         | 54±4                       | 10.4±0.5                          |
|             | 10                        | 23±3                       | 10.6±0.6                          |
| EMIMP99:1   | 0.5                       | 159±7                      | 11±2                              |
|             | 1                         | 132± 4                     | 8.2± 0.8                          |
|             | 2                         | 105±3                      | 7.7±0.8                           |
|             | 5                         | 59±6                       | 8.4±0.6                           |
|             | 10                        | 29±7                       | 7.9±0.8                           |

**Table S3:** Summary of the key performance parameters on different current density such as specific capacitance and ESR of EMIMP/CMC vs EMIMP/CNFc 99:1

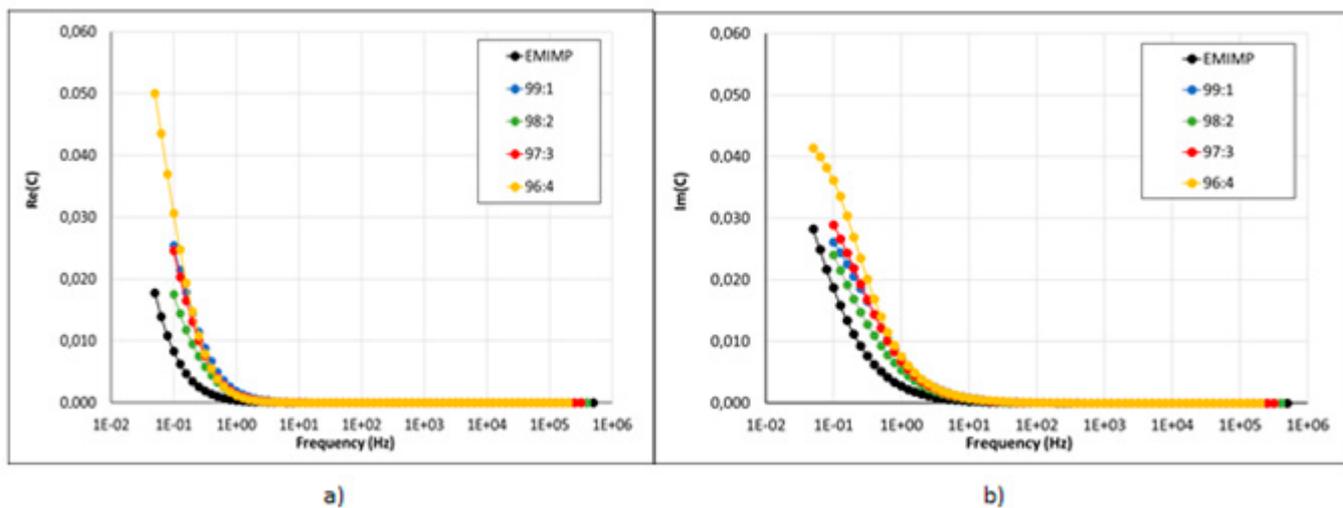
|           | Scan rate (mV s <sup>-1</sup> ) | $C_e$ (F g <sup>-1</sup> ) |
|-----------|---------------------------------|----------------------------|
| EMIMP     | 10                              | 2.4                        |
|           | 50                              | 0.6                        |
| EMIMP99:1 | 10                              | 6.9                        |
|           | 50                              | 1.1                        |
| EMIMP98:2 | 10                              | 8.1                        |
|           | 50                              | 1.1                        |
| EMIMP97:3 | 10                              | 8.3                        |
|           | 50                              | 1.2                        |
| EMIMP96:4 | 10                              | 7.3                        |
|           | 50                              | 1.1                        |

**Table S4:** Summary of specific capacitance of EMIMP and ionogels with different ratio of CNF<sub>c</sub> at 10 mV s<sup>-1</sup> and 50 mV s<sup>-1</sup>

scan rate for CV



**Figure S6:** Galvanostatic charge-discharge curve of EMIMP and ionogels with different ratio of CNFc at  $i = 1 \text{ mA cm}^{-2}$



**Figure S7:** a) Real part of the capacitance vs Frequency and b) Imaginary part of the capacitance versus Frequencies of EMIMP and ionogels with different ratio of CNFc