Supporting Information

3D-Printed Flexible Supercapacitors with Multi-Level Bonded Configuration via Ion Cross-linking.

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Calculations

Areal capacitance (C_A) , volumetric capacitance (C_V) and specific capacitance (C) of supercapacitors are calculated based on the galvanostatic charging and discharging curves according to the following equations:

$$C_A = \frac{I \times t}{\Delta V \times A} \tag{1}$$

$$C_V = \frac{I \times t}{\Delta V \times V} \tag{2}$$

$$C = \frac{I \times t}{\Delta V \times m} \tag{3}$$

where C_A , C_V and C are the areal, volumetric and specific capacitance, respectively. I is the discharge current, t is the discharge time, ΔV is the working voltage, A is the effective active area of the supercapacitor, V is the total volume of the supercapacitor and m is the mass loading of the total device. Areal energy density (E_A) and corresponding power density (P_A ,) of supercapacitors are calculated according to the following equations:

$$E_A = \frac{1000}{2 \times 3600} \times C_A \times \Delta V^2 \tag{4}$$

$$P_A = \frac{3600 \times E_A}{t} \tag{5}$$



Figure S1. a) Apparent viscosity as a function of shear rate for the MXene ink. b) The storage (G') and loss moduli (G") as a function of shear stress for MXene ink. c) Apparent viscosity as a function of shear rate for the Alginate-PAAm gel-like electrolyte.



Figure S2. a) SEM image of the sequentially bridged Ca-alginate/MXene electrode. b, c) TEM images of the sequentially bridged Ca-alginate/MXene electrode. Scale bars: a) 200 μ m, b, c) 50 nm.



Figure S3. a) XPS spectra of Ca-alginate/MXene, MXene/alginate, and MXene. b) XPS spectra C1s for MXene/alginate.



Figure S4. a) FTIR spectra for the Ca-alginate/MXene, and MXene. b) XRD patterns of Ca-alginate/MXene, MXene/alginate, and MXene.



Figure S5. Nyquist plots of CoC-M, ExC-M, and NonC-M supercapacitors.



Figure S6. The adhesive strength of the ExC-M supercapacitor.

Ref.	Capacitance	Deformations	Durability	Capacity retention
This work	2.7 F cm^{-2} (7 mA cm ⁻²)	Bending	1500 times	96.9%
1	$\begin{array}{c} (0.273 \text{ mF cm}^{-2}) \\ (300 \text{ mV s}^{-1}) \end{array}$	Bending	100 times	75%
2	3.3 mF cm ⁻² (0.02 mA cm ⁻²)	Bending	1200 times	92%
3	81.5 μF cm ⁻² (0.1 V s ⁻¹)	Bending	1800 times	87.5%
4	580 μF cm ⁻² (5.8 μA cm ⁻²)	Bending	1000 times	93%
This work	2.7 F cm ⁻² (7 mA cm ⁻²)	Stretching	2000 times	83.2%
5	216.2 mF cm ⁻² (10 mV s ⁻¹)	Stretching	1000 times	75%
6	450 mF cm ⁻² (1 mA cm ⁻²)	Stretching	1000 times	78%
7	23.2 F g ⁻¹ (0.5 A g ⁻¹⁾	Stretching	1000 times	81%

Table 1 The comparison of the flexibility, durability and electrochemical performance

 with literature reports on flexible supercapacitors.



Figure S7. a,b) Rheological behaviors of printable CNTs/alginate ink. a) Apparent viscosity in terms of shear rate for the CNTs/alginate ink. b) The storage (G') and loss moduli (G") as a function of shear stress for CNTs/alginate ink.



Figure S8. a,b) Rheological behaviors of printable Graphene/alginate ink. a) Apparent viscosity in terms of shear rate for the Graphene/alginate ink. b) The storage (G') and loss moduli (G") as a function of shear stress for Graphene/alginate ink.



Figure S9. a) and b) TEM images of Ca-alginate/CNTs electrode. Scale bars: a) 50 nm, b) 20 nm.



Figure S10. a) SEM image of the sequentially bridged Ca-alginate/graphene electrode. b,c) TEM and d) EDS mapping images of Ca-alginate/graphene electrode. Scale bars: a) 200 μ m, b) 500 nm, c) 20 nm, d) 200 nm.



Figure S11. a) charge-discharge curves of CoC-C supercapacitor collected at different current densities. b) Nyquist plots of CoC-C, ExC-C, and NonC-C supercapacitors. c) volumetric capacitances and d) specific capacitances of CoC-C, ExC-C, and NonC-C supercapacitors obtained at various current densities.



Figure S12. a) charge-discharge curves of CoC-G supercapacitor collected at different current densities. b) Nyquist plots of CoC-G, ExC-G, and NonC-G supercapacitors. c) volumetric capacitances and d) specific capacitances of CoC-G, ExC-G, and NonC-G supercapacitors obtained at various current densities.

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