Supplementary Information

Liquid Metal Dispersion by Self-Assembly of Natural Phenolics

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Section S1: Materials

Gallic acid (GA), sodium hydroxide (NaOH), lithium perchlorate (LiClO₄, 95.0%), and acetonitrile (99.5% ACN) were purchased from Sigma-Aldrich and used as received. Gallium ingots were purchased from Rotometals, Inc. Green tea bag (GT, Twinings) and cocoa nibs (Cacao Nibs, Macro) were purchased from a local market. High-purity (Milli-Q) H₂O with a resistivity of 18.2 M Ω .cm was obtained from an inline Millipore RiOs/Origin H₂O purification system.

Section S2: Characterisation

Scanning electron microscopy (SEM) images were obtained using a Nova Nano SEM 450 at an accelerating voltage of 10 kV. Energy-dispersive X-ray spectroscopy (EDS) mapping were obtained using a Nova Nano SEM 450 at an accelerating voltage of 20 kV. GA/Ga embedded paper samples sputtered with carbon coating were used for EDS analyses. Fourier transform infrared (FTIR) spectra were obtained using an attenuated total reflectance (ATR-FTIR) spectrometer (PerkinElmer). UV-Vis absorption spectra were recorded on a Cary 5000 (PerkinElmer) spectrophotometer. Transmission electron microscopy (TEM) images were obtained by high-resolution TEM (Phillips CM200, Amsterdam, Netherlands). X-ray photoelectron spectroscopy (XPS) was performed on an ESCALAB250Xi spectrometer (Thermo Scientific, UK). The incident radiation was monochromatic Al K α X-rays (1486.68 eV) at 120W (13.8 kV × 8.7 mA). Survey (wide) and high resolution (narrow) scans were taken at analyser pass energies of 100 eV and 20 eV, respectively. Base pressure in the analysis chamber was below 2.0 × 10⁻⁹ mbar. All data were processed using Advantage software and the energy calibration was referenced to the C 1s peak at 284.8 eV.

Electrical and electrochemical characterisation: The conductivity was calculated based on the resistance value obtained by the measurements through a two-probe technique using a digital multimeter (model Siglet SDM3065X). The equation for this calculation is as follows:

$$\sigma = \frac{L}{RA} \tag{1}$$

where L is the distance in between the probes, A is the transversal area and R is the resistance in Ω . Electrochemical assays were conducted to characterise the performance of the conductive paper as an electrode in the capacitor prototype. Cyclic voltammetry (CV) was carried out to assess the voltage window where a quasi-rectangular shape is obtained for the capacitor, considering different operational scan rates and define the current density limits applicable to the charge-discharge assays. CV of the control paper sample (i.e., without GA/Ga) was also conducted with a scan rate of 200 mV/s using 0.1M LiClO₄ in ACN as electrolyte solution.

Galvanostatic charge-discharge (GCD) cycles were obtained using different currents detailed in the text. GCD curves were also employed to evaluate the specific capacitance, *SC*, according to:

$$SC = \frac{\int I \cdot \Delta t}{\Delta V \cdot m_{Ga}}$$
 (2)

where I is the applied current, Δt is the time (s) of charge or discharge, ΔV is the difference between the potential (V) at the beginning and at the end of the charge or discharge, which in this case was fixed as 1 V and m is the mass (g) of Ga contained in the cellulose paper. The capacitance retention was analysed by GCD after 100 cycles applying 0.50 mA/g.

Electrochemical impedance spectroscopy (EIS) diagrams were taken at open-circuit cell potential (OCP) that is P over the frequency range of 100 kHz to 10 Hz. All experiments were performed using an CHI600e and CHI660e potentiostat/galvanostat workstation, at room temperature using 0.1M LiClO₄/ACN as the electrolyte solution.

Section S3: Synthesis of GA/Ga dispersion

In a typical synthesis, 20 μ L of liquid Ga (preheated at 60 °C on a hotplate) was added to a glass vial containing GA solution (2 mg mL⁻¹, 10 mL, pH adjusted to 6 by 1M NaOH, preheated at 60 °C on a hotplate). The vial was sealed and sonicated in a bath sonicator for 60 min and the bath temperature was maintained around 60 °C during sonication. After sonication the vial was taken out and left at room temperature for 2 min to allow the large particles to be settled down at the bottom of the vial. 1 mL of the supernatant suspension was taken out and washed 3 times in Milli-Q water via centrifugation (mini Spin plus, 8000 rpm, 2 min) to discard the excess reactants. The obtained particles were finally re-dispersed in Milli-Q water and this dispersion was used for further characterisation. Control experiments were also performed in Milli-Q water (without GA) in identical conditions as above. GA/Ga dispersions were also prepared with GA concentration of 0.5 and 5 mg mL⁻¹ in identical manner as above.

Section S4: Synthesis of GT/Ga dispersion

Boiling water was used for preparing the GT stock solution, briefly, 2 teabags were soaked in the boiling water (\sim 100 mL) for 3 min and the teabags were taken out. The resulting extract was cooled to room temperature and filtered through a 0.45 µm membrane filter to discard

large particles. The resulting filtrate (GT stock solution) was employed to prepare the GT/Ga dispersion, the process of which is identical to the process of GA/Ga dispersion as described in **Section S3**.

Section S5: Synthesis of Cocoa/Ga dispersion

Boling water was used for preparing the Cocoa stock solution. Briefly, 2 g of cocoa nibs was grinded with the aid of a mortar; the resulting powder was dispersed in boiling water and sonicated in a bath sonicator for 15 min. The resulting extract was cooled to room temperature and filtered through a 0.45 µm membrane filter to separate and discard large particles. The resulting filtrate (Cocoa stock solution) was utilised to prepare the Cocoa/Ga dispersion, the process of which was identical to the process of GA/Ga dispersion as described in **Section S3**.

Section S6: Impregnation of GA/Ga dispersion into paper membranes

The GA/Ga dispersion was prepared in identical conditions as described in **Section S3**. After sonication, the dispersion (10 mL) was washed 3 times in water via centrifugation and finally re-dispersed in 4 mL of water. 100 μ L drop of the final dispersion was spread over a commercial filter paper cut into the size of 2 cm × 7 cm (Whatman, pore size ~11 μ m, thickness of 0.2 mm). The last step was repeated until the entire dispersion was used up. The dimension of the paper was chosen to match the dispersion volume required to completely fill up the membrane matrix. Note that unlike **Section S3**, the large particles were not separated in this case. For quick evaporation of water, while spreading the drop, the paper sample was kept at 70 °C using a hot plate. After complete impregnation of the paper by GA/Ga particles, the composite paper was washed in Milli-Q water to remove loosely bound particles and dried. The dried composite paper was then sandwiched between two polypropylene sheet (thickness of 0.4 mm) and mechanically pressed by a press-rolling system (gap between rod 0.9 mm) to fuse the particles together in a matrix.

Section S7: Preparation of paper samples for electrochemical tests

GA/Ga impregnated paper samples after fusion (as obtained from Section S6) were cut into $4 \text{ mm} \times 6 \text{ mm}$ size and placed onto the surface of copper sheets of $4 \text{ mm} \times 20 \text{ mm}$ size. The joint was glued using Kapton[®] tape. The distance between the plates was 5 mm. The electrolyte used was 0.1 M LiClO₄/acetonitrile (ACN). A closed system was designed to avoid the ACN evaporation. The electrolyte solution was bubbled with N₂ for 10 min before each assay to remove the dissolved oxygen and prevent further oxidation of the electrodes.

Section S8: Supplementary Figures S1-16



Figure S1. Histogram showing the average particle size determined from SEM images of GA/Ga dispersion prepared using a GA concentration of 2 mg mL⁻¹.



Figure S2. SEM images of the GA/Ga dispersions obtained using GA concentrations of 0.5 (a) and 5 mg mL⁻¹ (b).



Figure S3. Histograms showing the average particle size determined from SEM images of GA/Ga dispersions obtained using GA concentrations of 0.5 (a) and 5 mg mL⁻¹ (b). (c) Plot showing the particle size variation with increasing GA concentration.



Figure S4. XPS survey spectra showing the elemental composition of the GA/Ga particles.



Figure S5. UV-Vis absorption spectra showing the difference of pure GA in 0.1 M HCl and GA adduct obtained after treating the GA/Ga particles in 0.1 M HCl.



Figure S6. (a) GT bag used for preparing the infusions. (b–d) Photographs showing the preparation of the GT/Ga dispersion. (e) SEM image showing the size and shape of the GT/Ga particles.



Figure S7. (a, b) Photographs showing the preparation of the Cocoa/Ga dispersion. (c) SEM image showing the size and shape of the Cocoa/Ga particles.



Figure S8. Histogram showing the average particle size determined from SEM images of GT/Ga dispersion.



Figure S9. Histogram showing the average particle size determined from SEM images of Cocoa/Ga dispersion.



Figure S10. EDS mapping (SEM) showing the presence of Ga in the GA/Ga impregnated paper membrane before pressing and fusion of the particles.



Figure S11. EDS mapping (SEM) showing the presence of Ga in the GA/Ga impregnated paper membrane after pressing and fusion of the particles.



Figure S12. Photograph showing the insulative nature (cannot power a light-emitting diode) of the GA/Ga incorporated paper membrane before pressing and fusion of particles.



Figure S13. (a-c) Experimental setup used for the electrochemical characterisation of the GA/Ga embedded conductive paper membranes after fusion.



Figure S14. (a) Specific capacitance of the composite paper after fusion as a function of current density. (b) CV plots showing the stability of the composite paper (red trace) and paper membrane without GA/Ga (control, grey trace) using 0.1 M LiClO₄/ACN electrolyte solution

with a scan rate of 200 mV/s. (c) Analysis of electroactivity of the composite paper by GCD plot during the first 100 cycles by applying 0.50 mA/g.



Figure S15. GCD plot of specific capacity vs potential for the composite paper after fusion, the best performance achieved at 0.28 mA/g in the potential window from -0.50 to 0.50 V.



Figure S16. EIS assays for the GA/Ga embedded conductive paper membranes: (a) Nyquist plot with an inset corresponding to the amplified area of the low frequency zone for better visualisation and Bode plots (b, c) of the composite paper-based capacitor.

It is evident from the EIS assays that a diffusion-limited process drives the kinetics of the capacitor. The Nyquist and Bode plots are displayed in Figure S16. A straight line was observed in the Nyquist plot (Figure S16a) which is consistent with the trend of a typical electrical double-layer of porous capacitors.¹ The inset (Figure S16a) shows that at high frequencies, the

semicircle characteristic of the charge transfer resistance was absent, meaning that our process was improved by the porous structure of the cellulose paper electrode and possibly the presence of GA adduct released after shell rupture during fusion, which favoured the permeation of the electrolyte and enhances the ion transfer kinetics.² The shapes of the Bode plots as shown Figure S16 (b, c) are typical of conducting materials. However, the linear trend observed around 2×10^2 Hz at the centre of the Bode-phase plot indicates a diffusion limited process at the electrolyte interface that agrees well with the trend observed for the Nyquist plot.

Section S9: References

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