

## Supporting information

### Accounting for carbon black

Due to the high percentage of carbon black (CB) in the electrodes, a 9:1 CB:PVDF electrode was used as a control to isolate and understand the contribution of both the CB and the active components of the electrodes. The reversible capacity of the CB electrode after the first 100 cycles was 192 mAhg<sup>-1</sup> (Figure S1). The capacity retention of the CB electrode was ~100% over 100 cycles, suggesting that any capacity loss seen in the electrodes is likely to be entirely from the active component.

Assuming a linear capacity-ratio relationship and that the PVDF is non-active, the CB contribution to the copper tartrate electrodes was determined to be 78 and 69 mAhg<sup>-1</sup> for the as purchased and acid-formulated electrodes respectively (85 mAhg<sup>-1</sup> when calculated based on the mass of the acid). This is 37/91 and 33/92 of the capacity of the CB electrode (accounting for the amount of carbon black in the two electrodes respectively). The theoretical capacity values were then calculated by subtracting the CB contribution from the electrode capacities, and then multiplying the residual capacity by the ratio of active/Cu TAR, where "active" is CB + Cu TAR, to simulate a wholly Cu TAR electrode. The number of Li per copper tartrate is then calculated as follows:

$$\frac{\text{Theoretical capacity} * \frac{3600}{1000} * MW}{F}$$

Where *MW* is the molecular mass of copper tartrate and *F* is Faraday's constant.

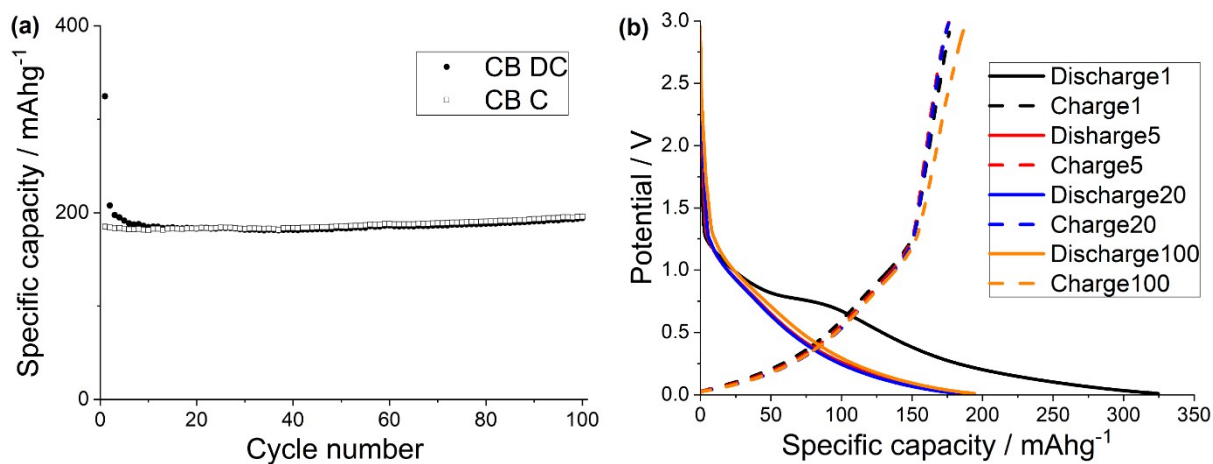


Figure S1: (a) Discharge (filled) and charge (non-filled) specific capacity vs cycle number for carbon black. (b) 1<sup>st</sup> (black), 5<sup>th</sup> (red), 20<sup>th</sup> (blue) and 100<sup>th</sup> (orange) discharge (solid) and charge (dashed) potential vs specific capacity curves for the carbon black electrode.

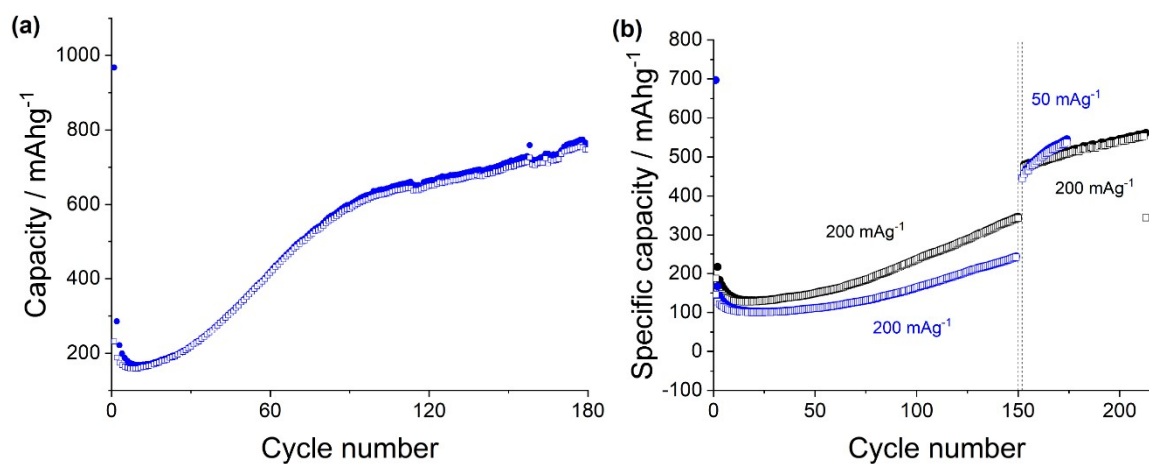


Figure S2: (a) Discharge (filled) and charge (non-filled) specific capacity vs cycle number of the 180 pre-cycles for the rate capability experiment, cycling at 50 mA g<sup>-1</sup> between 0.01 and 3 V. (b) Discharge (filled) and charged (non-filled) specific capacity vs cycle number for as purchased copper tartrate electrode of the 150 pre-cycles for the rate capability experiments, with cycling at 200 mA g<sup>-1</sup> between 0.01 and 3 V. Dashed lines represent where the rate performance experiment was performed, and following cycles were at 50 mA g<sup>-1</sup> (blue) and 200 mA g<sup>-1</sup> (black).

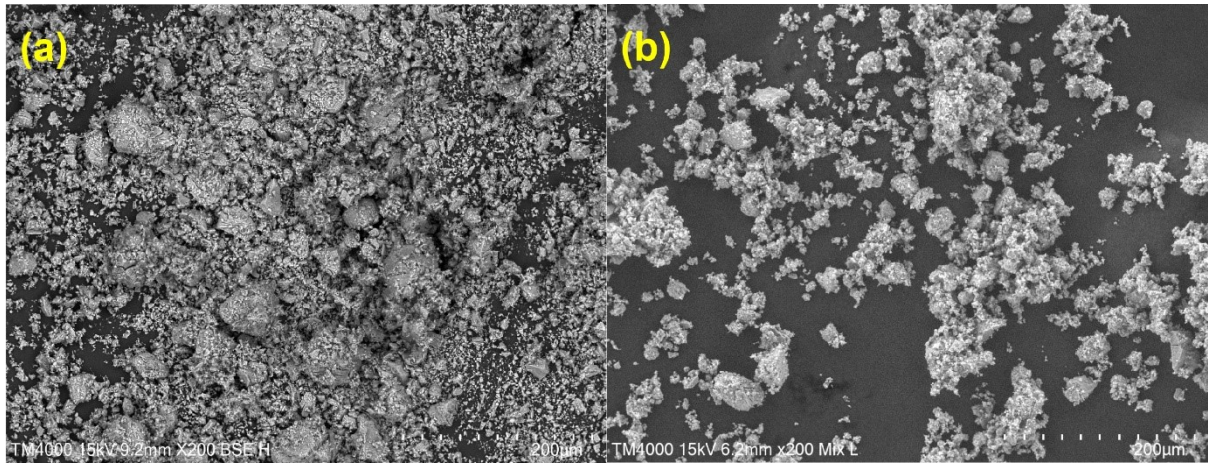


Figure S3: (a) SEM (200x, 15 kV) of copper tartrate as purchased after grinding with mortar and pestle. (b) SEM (200x, 15 kV) of copper tartrate after milling.

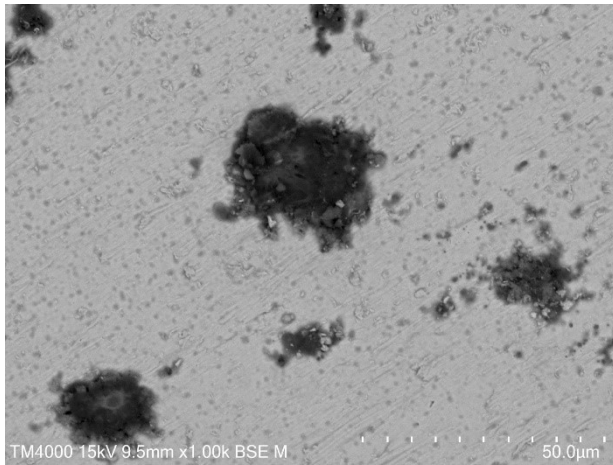


Figure S4: SEM (1000x, 15 kV) of the copper substrate from the copper tartrate electrode after 250 cycles.

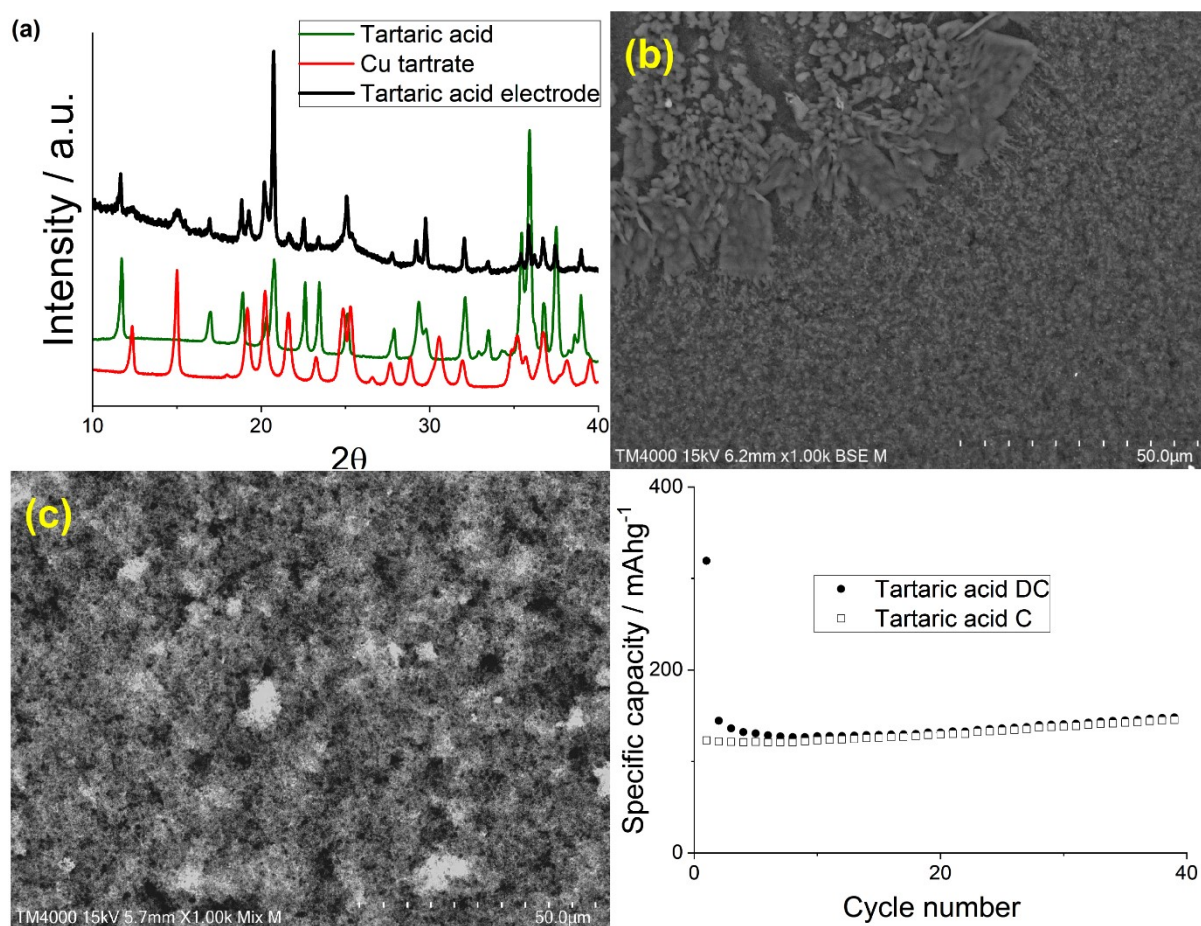


Figure S5: (a) XRD patterns of copper tartrate (red), tartaric acid (green) and partially reacted tartaric acid electrode (black). (b) SEM (1000x, 15 kV) of partially reacted tartaric acid electrode. (c) SEM (200x, 15 kV) of carbon black electrode. (d) Discharge (filled) and charge (non-filled) specific capacity vs cycle number for tartaric acid electrode, cycling between 0.01 and 3 V at 50 mAhg<sup>-1</sup>.

USANS background subtraction

It is important to note that USANS measurement was conducted down to the  $q$  range of  $3.6 \times 10^{-5} \text{ \AA}^{-1}$ .

However, further analysis of the copper substrate revealed that it scattered anisotropic scattering below  $2 \times 10^{-4} \text{ \AA}^{-1}$  (Figure S6a). Hence, subtraction of the copper background was complicated, as the samples were not spun during data collection, nor were the samples mounted at the same orientation. Each sample had both the 0 degree and 90-degree copper background subtracted from it and it was determined that scattering above  $2 \times 10^{-4} \text{ \AA}^{-1}$   $Q$  range was not affected by this anisotropy (Figure S6b). For the as purchased, the scattering below  $2 \times 10^{-4} \text{ \AA}^{-1}$  was dominated by the electrode, making the difference non-significant. For the milled electrode, use of the 90-degree background

resulted in nonsensical data including negative intensities. The 0-degree background demonstrated no significant change in the slope, making it the best choice. For the acid formulated and carbon black electrodes, the samples appear to have been run at an angle somewhere between 0 and 90 degrees, as both subtractions produced anomalous results. Hence, all data points below  $2 \times 10^{-4} \text{ \AA}^{-1}$  were excluded.

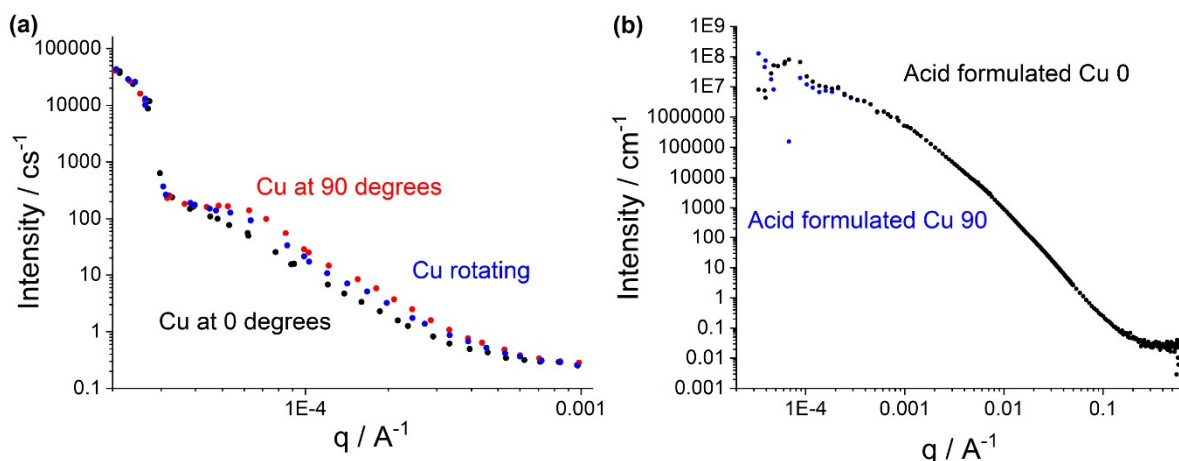


Figure S6: (a) Raw USANS data showing the anisotropic scattering of the copper foil substrate. Measurements were taken from the sample at 0 degrees (black), 90 degrees (red) and rotating (blue). (b) Comparison of U/SANS data for the acid formulated electrode using the 0 degrees (black) and 90 degrees (blue) copper backgrounds. Complete overlap of data points is achieved by  $q > 2 \times 10^{-4}$  demonstrating no anisotropic scattering above this  $q$  range.

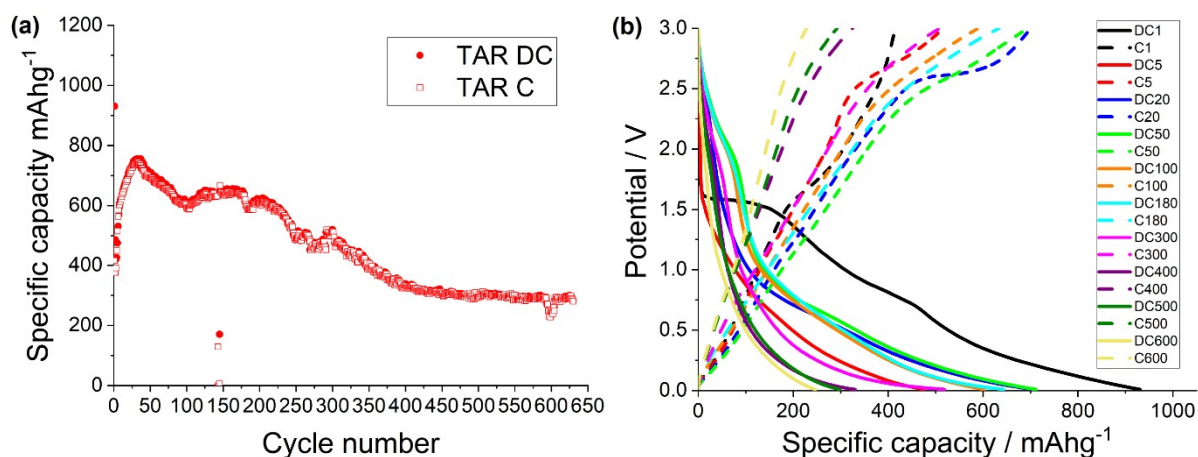


Figure S7: (a) Discharge (filled) and charge (non-filled) specific capacity vs cycle number for tartaric acid formulated electrode. (b) 1<sup>st</sup> (black), 5<sup>th</sup> (red), 20<sup>th</sup> (blue), 50<sup>th</sup> (green), 100<sup>th</sup> (orange), 180<sup>th</sup> (cyan), 300<sup>th</sup> (magenta), 400<sup>th</sup> (purple), 500<sup>th</sup> (dark green) and 600<sup>th</sup> (yellow) discharge (solid) and charge (dashed) potential vs specific capacity curves for acid formulated electrode.