**Electronic Supporting Information**

**Laser-induced highly oriented pyrolytic graphite for high-performance screen-printed electrodes**

Alejandro Fidel Alba, Joseba Totoricaguena-Gorriño, Lia Campos-Arias, Nikola Peřinka, Leire Ruiz-Rubio, José Luis Vilas-Vilela, Senentxu Lanceros-Méndez, Francisco Javier del Campo.

**Table of Contents**

| List of laser conditions and resulting in-plane crystallite size as measured from Raman spectra. | Table ESI1 |
|---|---|
| Electrode surface colour as a function of applied laser energy. | Figure ESI1 |
| Electrode resistance as a function of applied laser energy. | Figure ESI2 |
| Printed graphite ablation depth as a function of laser energy. | Figure ESI3 |
| Thermogravimetry and Differential Scanning Calorimetry of a graphite paste sample. | Figure ESI4 |
| Water-in-air contact angle as a function of applied laser energy. | Figure ESI5a and Figure ESI5b |
Correspondence between laser power and applied energy

Table ESI1. Correspondence between the laser power settings and the applied energy density considering the work condition, as well as determination of in-plane crystallinity parameter $L_a$ as a function of applied energy, according to Raman spectroscopy.

| LASER POWER SETTING (% OF MAX) | APPLIED LASER POWER (W) | ESTIMATED APPLIED ENERGY DENSITY (mJ cm$^{-2}$) | RAMAN $I_D/I_G$ | IN-PLANE CRYSTALLINITY PARAMETER $L_a$ (nm) |
|-------------------------------|-------------------------|-----------------------------------------------|-----------------|------------------------------------------|
| 0                             | 0                       | 0                                             | 0.866           | 19                                       |
| 6                             | 1.8                     | 5.1                                           | 0.698           | 24                                       |
| 7                             | 2.1                     | 6.0                                           | 0.497           | 34                                       |
| 8                             | 2.4                     | 6.8                                           | 0.159           | 105                                      |
| 9                             | 2.7                     | 7.7                                           | 0.145           | 116                                      |
| 10                            | 3.0                     | 8.5                                           | 0.126           | 133                                      |
| 11                            | 3.3                     | 9.4                                           | 0.149           | 112                                      |
| 12                            | 3.6                     | 10.2                                          | 0.069           | 243                                      |
Change in colour intensity as a function of applied laser power

Figure ESI1. Left. Colour intensity distribution (histogram) measured by ImageJ from 1200 dpi 1x1 electrode images. The figure on the left represents the histograms for each electrode. The square symbol represents the mode value and the bars represent the standard deviation from the mean values. Colour intensity is calculated (ImageJ) in a scale between 0 (black) and 255 (white). The right-hand side figure represents the change as % of the ready-printed electrode value.

While electrode colour may not convey useful quantitative information on the material itself, it can be used as a straightforward means to control the process (repeatability) and the quality of the electrodes. Above 7.7 mJ cm$^{-2}$ the electrode is thin enough to let some light through it and reflect on the white background of the flatbed scanner.
Effect of laser power on electrode electrical resistance

Figure ESI2. Electrical resistance of the lasered electrodes as a function of applied power.

The resistance increases linearly up to 7.7 mJ cm$^{-2}$, consistent with the thinning of the printed layer due to material ablation. Above 7.7 mJ cm$^{-2}$ the electrodes become much more resistive, presumably because of discontinuities in the film arising from the laser ablation process.
Ablation depth

**Figure ESI3.** Thickness of graphite screen-printed coating after being exposed to different laser powers. Laser ablation removes material at an approximate rate of 1.9-2.2 µm thickness per mJ cm$^{-2}$ applied. This is equivalent to an ablation rate of 0.19-0.22 mm$^3$ J$^{-1}$. 
Thermogravimetric analysis and Differential Scanning Calorimetry of a carbon paste sample

Figure ESI4. Thermogravimetric analysis (TGA, black line, left axis) and differential scanning calorimetry (DSC, red line, right axis). TGA shows that solvent residues evaporate up to about 200°C. The binder, which accounts for 20-25% of the starting mass, decomposes up to 420-450 °C. Above this temperature only carbon (graphite) remains, which decomposes in several steps, presumably lower quality graphite is consumed first and, above 800°C there is nothing left. The DSC peaks show the transition temperatures where the various decomposition events occur.
Surface tension variations

Figure ESI5a. Contact angle of a water droplet on the surface of a graphite paste coating following laser treatment at different applied energy.
Figure ESI5b. Changes in contact angle as a function of applied power. The square dots represent the average of 5 measurements, and the error bars show the corresponding standard deviations.