

Supplementary information

Figure A shows the change in plasma shape. Observationally, this change of plasma discharge regime takes about 5 min and is noted as an intermediate plasma regime. At 20 min, the filamentary plasma regime is established.

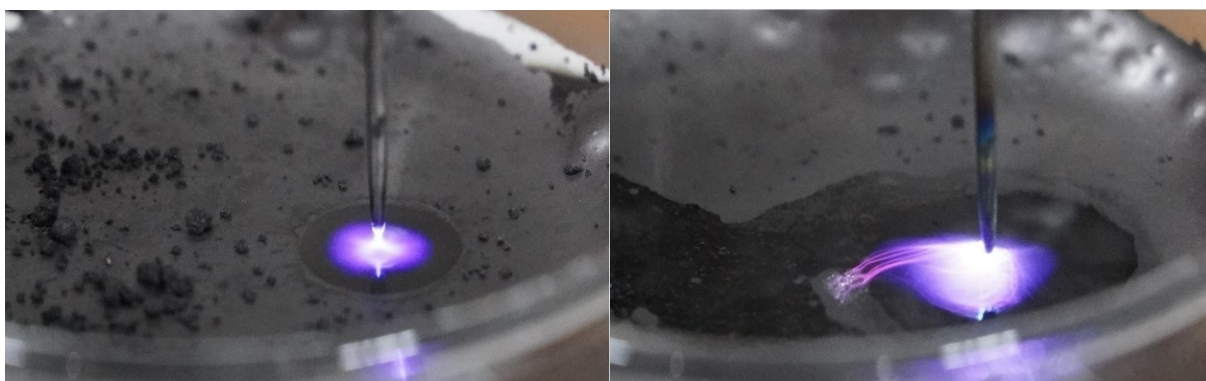


Figure A: images of the plasma discharges at the liquid surface with KNG 180 samples: (left) image exhibits a columnar plasma taken 5 min after the beginning of the treatment and (right) after 40 min with a transition to a filamentary plasma.

Figure B presents the pulse power and the mean power evolution for the second process (200 mg of KNG 180; 40 mL of demineralized water; 5 g.L^{-1}). The powers present a rapid increase in the first ten minutes of process, and reach values of 20 kW and 120 W for pulse power and mean power, respectively. The maximum mean power reaches a maximum value of 147 W at 20 minutes of process.

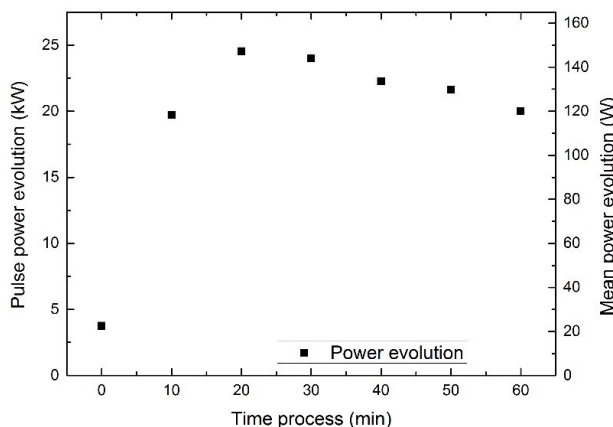


Figure B: pulse power and the associated mean power evolutions during the plasma treatment process of 200 mg of KNG 180 in 40 mL of water and in air.

Figure C shows normalized X-ray diffraction pattern of pristine KNG 180 and treated samples. The most intense peak around 12° is assigned to (002) reflection of hexagonal graphite ($a = 246 \text{ pm}$, $c = 671 \text{ pm}$), and a set of small diffraction peaks around 20° correspond to hkl reflection of hexagonal graphite too.

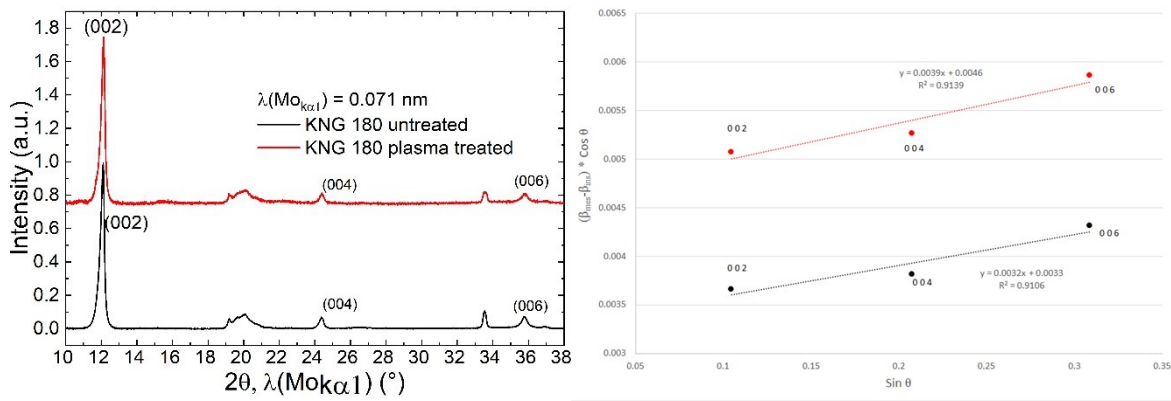


Figure C: X-ray diffractograms ($\lambda(\text{MoK}\alpha 1) = 0.70930 \text{ \AA}$) and the Williamson-Hall plot of the KNG 180 untreated sample (black) and treated by plasma (red).

Figure D and E show the decomposition of the Raman spectra obtained. Raman spectroscopy is a common technique for the analysis of carbon materials. Four major features can usually be observed for graphite and graphene:

- the D band (around 1350 cm^{-1}) corresponding to the A_{1g} vibration mode. This mode is forbidden in the case of graphite or graphene single crystals. It and only appears for a polycrystalline material or at the edge of a particle;
- the G band (around 1580 cm^{-1}) is attributed to the E_{2g} mode;
- the D' band (around 1620 cm^{-1}) related to the D band;
- the 2D band (around 2700 cm^{-1}) which is an overtone of the D band. It is always active and related to the stacking order of the material. For a single layer of graphene, the 2D band is a single sharp Lorentzian peak. For a few-layer graphene sample, the number of components varies with the number of layers, and above five layers, the 2D band becomes not distinguishable from the graphite one, which presents two components. In the case of turbostratic graphite, because of the electronic decoupling of the layers due to their random stacking, the 2D band is a single Lorentzian peak, as for graphene, but broadened

Curve fitting was performed on the $1200\text{-}1700 \text{ cm}^{-1}$ region using Lorentzian curves for all the bands. It confirms the presence of the D, G and D' bands. The 2D band can be fitted with three Lorentzian components; two components were attributed to the hexagonal graphite and one to the turbostratic graphite. The used software was "origin"

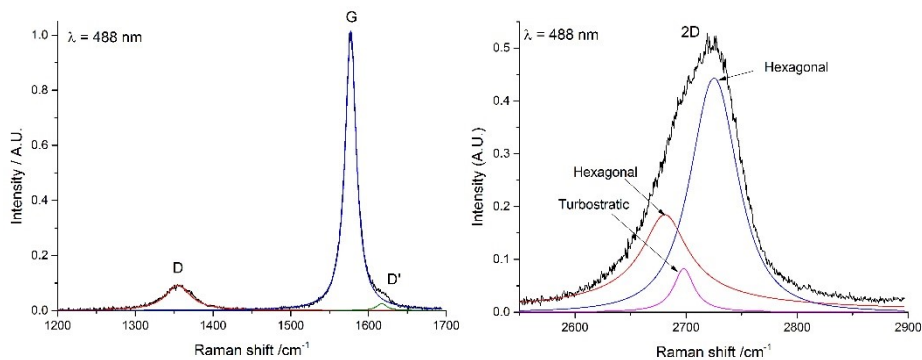


Figure D: decomposition of the Raman spectra of the untreated sample

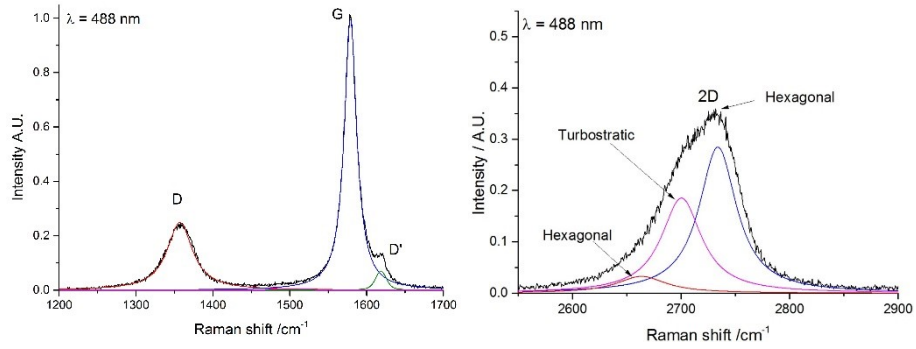


Figure E: decomposition of the Raman spectra of the treated sample by plasma

Figures F and G show the thermograms and mass spectrometry signals for untreated and plasma treated KNG 180 samples.

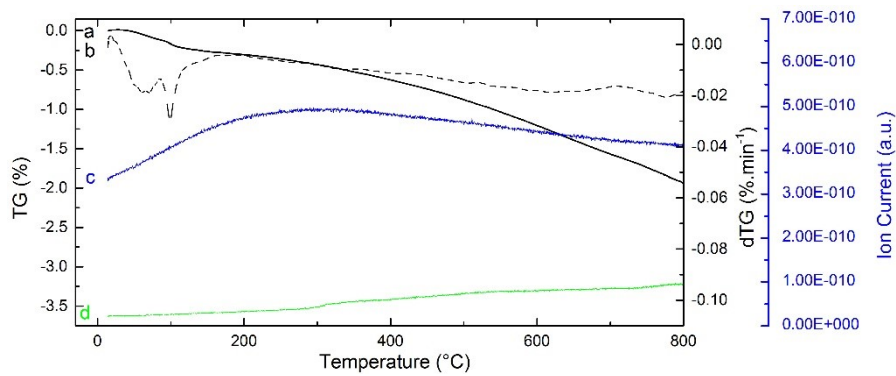


Figure F: thermogram (a), differential thermogram (b) and mass spectrometry signals for the carbon monoxide ($m/z=28$) (c) and carbon dioxide ($m/z=44$) (d) for the untreated KNG 180.

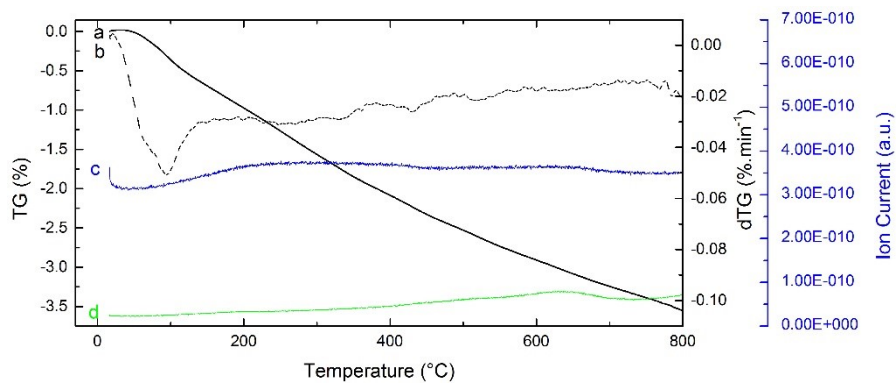


Figure G: thermogram (a), differential thermograms (b) and mass spectrometry signals for the carbon monoxide ($m/z=28$) (c) and carbon dioxide ($m/z=44$) (d) for the KNG 180 plasma treated.