

The effect of hydrogen plasma on the chemical and structural modification of graphene

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Abstract

Graphene based devices fabrication is currently impeded by its metallic behavior and its lack of reactivity making it hard to create graphene based transistors or sensors. In order to overcome this problem, various approaches to tune and control the electronic properties of graphene have been considered. Among them, plasma treatment has been shown to be a very interesting and controllable way of chemically modifying graphene.[1-3] Furthermore, it has the advantage over other processes to be easily implemented in the electronic device production line.

In this work, we investigated the controlled modification of graphene using hydrogen (H₂) rf-plasma treatment.[1,3] Structural changes induced on the graphene flakes are first monitored using Raman spectroscopy.[2,3] It is found that changing the plasma parameters (i.e. pressure, power, exposure time and position of the sample in the plasma chamber) allows fine tuning of the modification induced on the graphene. For example, figure 1 shows the Raman spectra of monolayer graphene for different exposure time (figure 1a) and different positions of the sample inside the plasma chamber (figure 1b). We can see that these two parameters affect greatly the amount of defects induced on the graphene. In addition, it is noticed that the hydrogen pressure is another particularly important parameter to take into account.

Defect formation kinetics under plasma is also found to vary with the number of graphene layers (see figure 1c). Surprisingly, at specific conditions, bilayer and trilayer graphene seem to be more reactive than monolayer graphene (higher intensity and larger FWHM of the D peak are recorded) and layer-by-layer thinning of multilayer graphene is occurring.

In order to support the Raman results, complementary measurements performed using micro-focused X-ray photoelectron spectroscopy (micro-XPS) and atomic force microscopy (AFM) are performed. The first technique give us information on the chemical state and the carbon content of mono and bilayer graphene after hydrogenation, while the latter allows visualization of the structural damages induced on the graphene sheets.

In conclusion, we carefully studied the hydrogenation of graphene inside a low temperature plasma. We show that graphene modification is strongly dependent on the plasma parameters (pressure, power, exposure time and position of the sample) as well as on its number of layers. This work sheds light to the complex process that is a hydrogen plasma which allows in turn controlled tuning of the functionalization of graphene.

References

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Figures

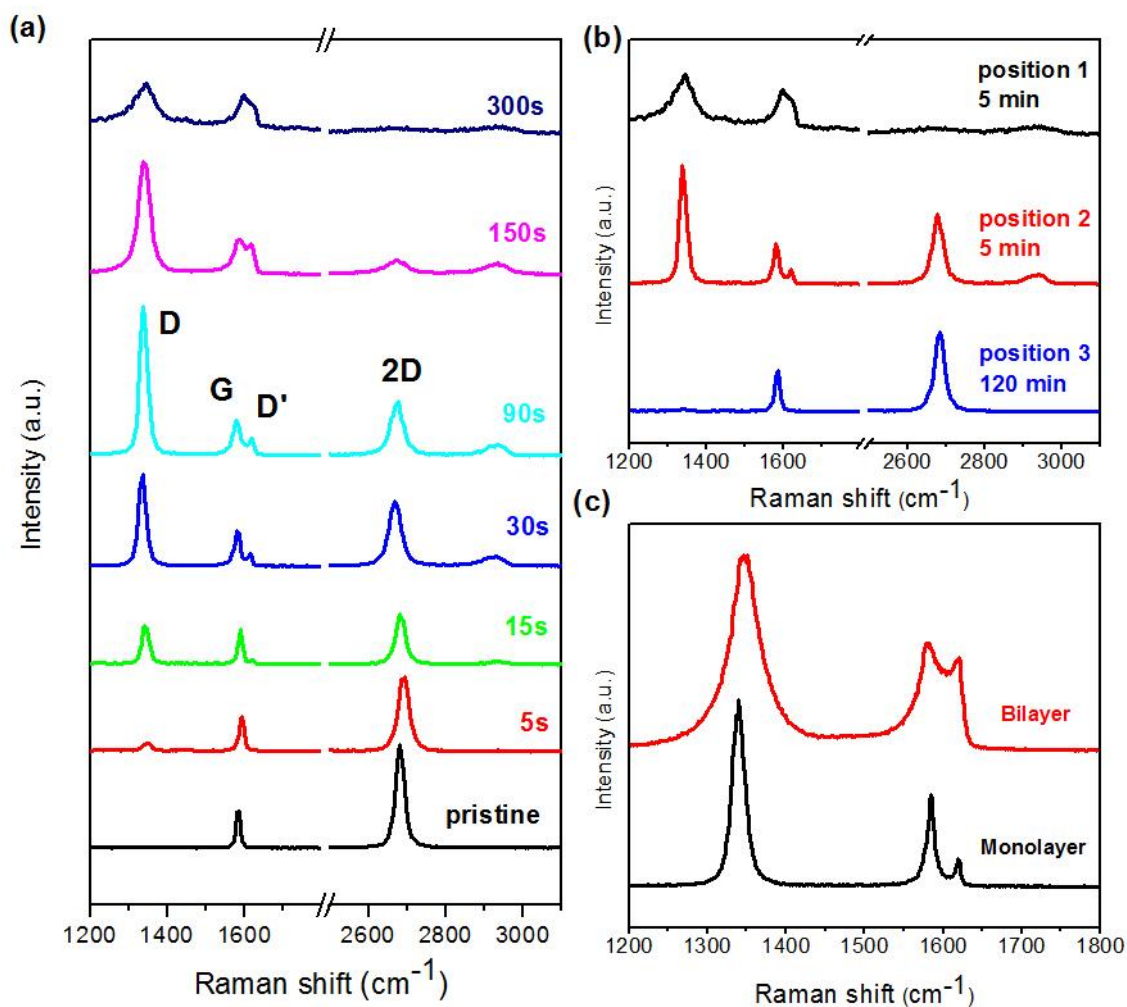


Figure 1. (a) Evolution of Raman spectra of monolayer graphene upon increasing hydrogen plasma exposure. (b) Raman spectra of monolayer graphene modified under hydrogen plasma placed at different positions in the plasma chamber. Positions 1, 2 and 3 correspond respectively to inside the discharge, outside the discharge (20 cm away from position 1) and further away from the discharge (40 cm away from position 1). (c) Raman spectra of mono- and bilayer graphene modified at the same time under the same plasma conditions.