Synthesis of carbon materials and hydrogen in a plasma fluidized bed reactor

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Highlights

- A non-thermal plasma process is developed for carbon nanomaterials synthesis.
- High purity graphene was produced from a mixture of methane in argon.
- Effect of a fluidized bed of particles is investigated.

1. Introduction

Non-thermal plasma is a promising technology for high purity carbon materials synthesis from hydrocarbons in a fast, flexible and adjustable process. More specifically rotating gliding arc (RGA) is a promising plasma source due to its characteristics in both equilibrium and non-equilibrium conditions [1]. In this study, graphene synthesis is carried out in a rotating gliding arc reactor with and without a fluidized bed of particles at atmospheric pressure. The effects of the particles' addition on the structures and properties of the carbon materials produced is explored and compared with the results of plasma in an empty reactor.

2. Methods

The tests were performed in a stainless-steel fluidized bed RGA reactor, with and without particles, as presented in Figure 1. The reactor consists of a cylindrical outer electrode with a diameter of 32 mm, whereas the high voltage electrode is a truncated cone with a diameter of the bottom and top base of 28 and 13 mm respectively, and a height of 30 mm. The narrowest gap between the two electrodes is 2 mm. The gas is supplied radially via twelve tangential gas inlets placed at an angular distance of 30 degrees near the bottom plane and axially via a second gas inlet consisting of forty-nine vertical tubes with an inner diameter of 1 mm drilled in the cone. On top of this, two stainless steel inserts equipped with trapezoidal baffles are positioned at 10 mm distance. The fluidized bed consist of 3 g of γ-alumina particles in the range 80 – 150 μm.

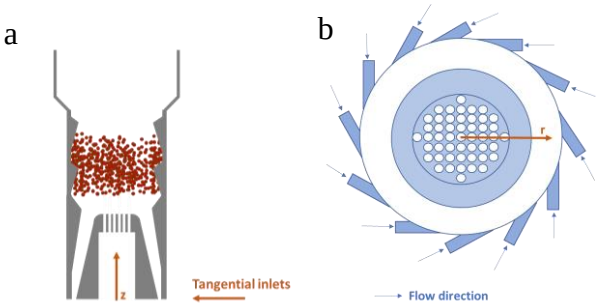

Figure 1. Schematic of the rector employed in the experiments: **a)** front view and **b)** top view.

3. Results and discussion

To investigate the carbon materials synthesis, a mixture of 8 % methane in argon was fed to the reactor (total flowrate 15 l/min). a methane conversion of 11.8 % was obtained in the case of empty reactor, with a 14.5 % and 85.1 % selectivity towards acetylene and hydrogen respectively. The carbon deposited on the high voltage electrode and on the ground electrode was collected and characterized. The results are presented in the figures below.

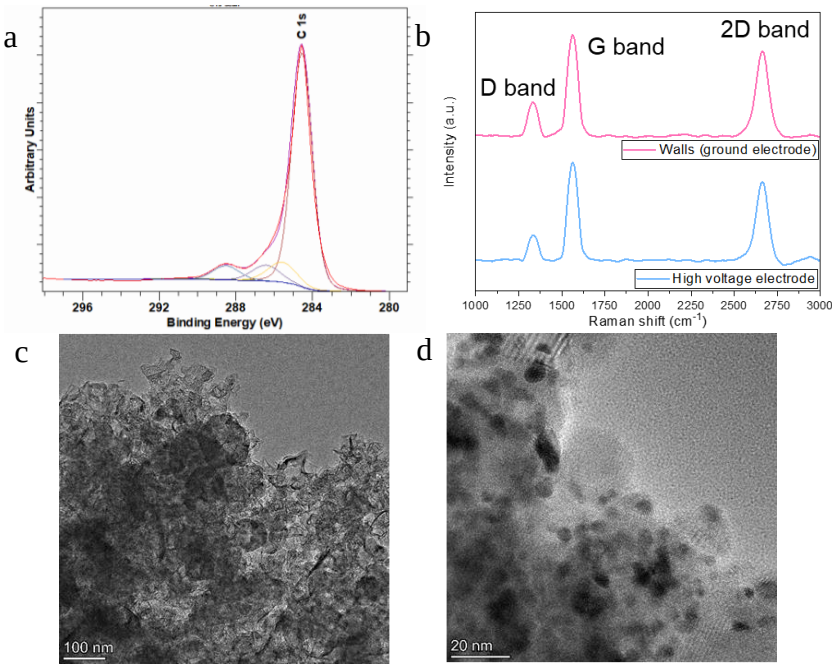

Figure 2. a) XPS spectrum, **b)** Raman spectrum, **c)** low-magnification image and **d)** high magnification image for the empty reactor tests.

The surface composition of the produced carbon materials was obtained by XPS technique. The deconvoluted C1s peak show five binding energies of 284.50, 285.5, 286.30, 288.7, and 290.7 eV, representing C–C (sp2), C–C (sp3), C–O, C=O groups and π-π* satellite bonds, respectively. The first and main contribution at 284.5 eV can be assigned to the formed graphitic structure carbon. This peak clearly indicates that the synthesized materials possess predominantly sp^2 bonds. The relative content of sp^2 carbon in the sample was calculated as the ratio of the corresponding peak area over the total C 1s peak area and it was found to be approximately 75 % [2]. The Raman spectra of the samples show a major peak at around 1580 cm^{-1}, assigned to the G band. The peak is due to E_{2g} stretching mode of sp^2-bonded carbon in a 2D hexagonal lattice. By contrast, the band at around 1330 cm^{-1} is assigned to the disorder-induced D band, related to the presence of defects, such as vacancies or non-hexagonal rings [3]. The ratio of D and G peak intensity (I_D/I_G) is associated with graphite crystallization degree, which is expected to be lower than 0.6 for a high graphite degree with few defect structures (our case 0.25) [4]. The study of the structure of the carbon material was conducted using high-resolution TEM. Macroscopically, the graphene is wrinkled and appears clumped consistently through multiple samples [5].

4. Conclusions

Graphene was successfully produced with very high selectivity in a rotating gliding arc reactor. Overall, the gliding arc discharge presents the potentials in the nanomaterial synthesis with high productivity and high purity.

References

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Keywords
Plasma, graphene, rotating gliding arc, fluidized bed