Study of the influence of thermal treatment on multi-layer graphene samples obtained by solvothermal reaction: improvement of the structural characteristics

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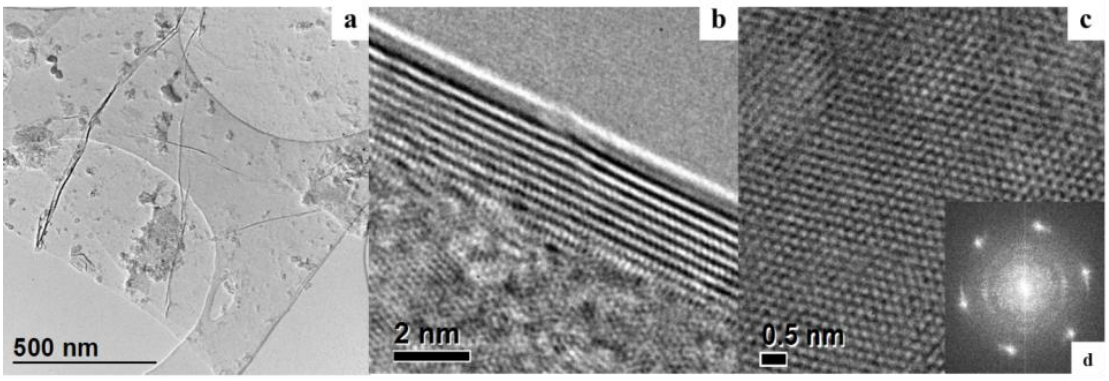
Abstract

Graphene has become one of the most studied carbon materials. It exhibits exceptional properties: the highest charge carrier mobility and thermal conductivity reported, great mechanical properties, almost complete transparency, perfect impermeability and high surface area. These features open a wide range of promising applications and several methods of synthesis have been developed. Micromechanical cleavage, liquid phase exfoliation of graphite and its intercalation compounds, supported growth on metal surfaces and silicon carbide represent the current major techniques for graphene fabrication [1]. In spite of the intensive research focused on these different methods, none yields industrial quantities of high-quality graphene. Then, alternative ways have been proposed and we focused on solvothermal synthesis. A pioneer study reported the synthesis of graphene from solvothermal reaction between ethanol and sodium followed by a flash pyrolysis [2] and later work modified the thermal treatment by replacing it by combustion in air [3]. Here we report the first study of the variation of the parameters of the thermal treatment. We performed solvothermal reaction between ethanol and sodium at 220°C during 72 h with a pressure of 200 bar. The reaction yields metastable and air-sensitive sodium ethoxide assumed to adopt a clathrate-like structure with trapped ethanol molecules. This compound is then pyrolyzed at 20°C.min^{-1} under nitrogen flow with different conditions: at 800°C, 825°C and 850°C during 4 h or 24 h. The final carbon materials were characterized using various techniques (TEM, XRD, TGA under dry air, TGA-MS under helium, Raman spectroscopy and nitrogen adsorption). They revealed the production of multi-layer (below 10 layers) graphene powders with specific surface areas reaching the theoretical limit (from 1500 m^2.g^{-1} to 3000 m^2.g^{-1}) and exhibiting clean surfaces, exempt of heteroatoms and showing not many surface functions. This study shows the strong influence of temperature and time of pyrolysis on purity, crystallinity and thickness of the samples, and enables to go toward the control of these characteristics. Post-synthesis vacuum annealing is proposed to still improve the structural quality, and several applications are envisaged, for example integration in fuel cells after alkali metals intercalation and dispersion.

References

[1] K. S. Novoselov, V. I. Fal'ko, L. Colombo *et al*., Nature, **490** (2012) 192-200.
[2] M. Choucair, P. Thodarson, A. Stride, Nature nanotechnology, **4** (2009), 30-33.
[3] S. M. Lyth, H. Shao, J. Liu, K. Sasaki, E. Akiba, International Journal of Hydrogen Energy **39** (2014) 376-80.

Figures



TEM micrographs (incident beam 80 keV) of the sample pyrolyzed at 825°C during 24h: (a) low resolution, (b) and (c) high resolution, (d) FFT of (c).