

Electron Microscopy of Graphene Materials Produced by Combustion Synthesis

Khachatur V. Manukyan¹, Sergei Rouvimov², and Alexander S. Mukasyan^{1,2}

1. Department of Chemical and Biomolecular Engineering, University of Notre Dame, Notre Dame, IN, 46556, USA

2. Notre Dame Integrated Imaging Facility (NDIIF), University of Notre Dame, Notre Dame, IN, 46556, USA

Graphene, atomically thin carbon sheets, and its structural derivatives (bi- or few-layer graphene) have attracted great attention due to their exceptional charge transport, thermal, optical and mechanical properties [1-3]. We developed combustion synthesis (CS) method for hybrid structure of *graphene sheets* and *porous carbon nanoparticles* (Figure 1a) by a novel exothermic *self-sustaining* reaction between a refractory silicon carbide (SiC) and polytetrafluoroethylene (PTFE) under the *inert atmosphere*. The combustion synthesis (CS) of graphene is confirmed by both transmission electron microscopy and Raman spectroscopy. The mechanism for CS of graphene is also suggested. It is shown experimentally that the tetrafluoroethylene (C_2F_4) gas, released due to PTFE decomposition in the combustion wave, reduces SiC to tetrafluorosilane (SiF_4) gas (Figure 1b) and disordered carbon particles with folded “native” graphene layers on their surfaces (Figure 1c). The continuous supply of carbon, in the form of C_2F_4 , enables further rapid growth of “free-standing” graphene flakes on the surface of those graphene-coated porous particles.

High Resolution Transmission Electron Microscopy (HRTEM) images clearly show that flakes appear to be single layer (Figure 1d) or few (Figure 1d) layers of graphene. The specific surface area of the reaction product is $\sim 450\text{m}^2/\text{g}$, which is hundred times higher from surface area of initial reactants. This significant difference is related to formation of meso-pores (~ 3.5 nm in size) during the reaction owing to the removal of silicon from the SiC lattice. The unique aspect of reported method for synthesis of graphene materials does not require external energy source, as it occurs in a self-sustained synergetic manner. This method is flexible in terms of tuning the synthesis conditions, which provide materials with different microstructure and composition. It is also important that the low oxygen content in the reacting system allows synthesis of oxygen-free products. The hybrid structure of *graphene sheets* and *porous carbon nanoparticles* combine the merits of electrical properties of graphene and large specific surface areas for porous carbon, and is a desirable structure for applications including batteries, super-capacitors, and catalysis.

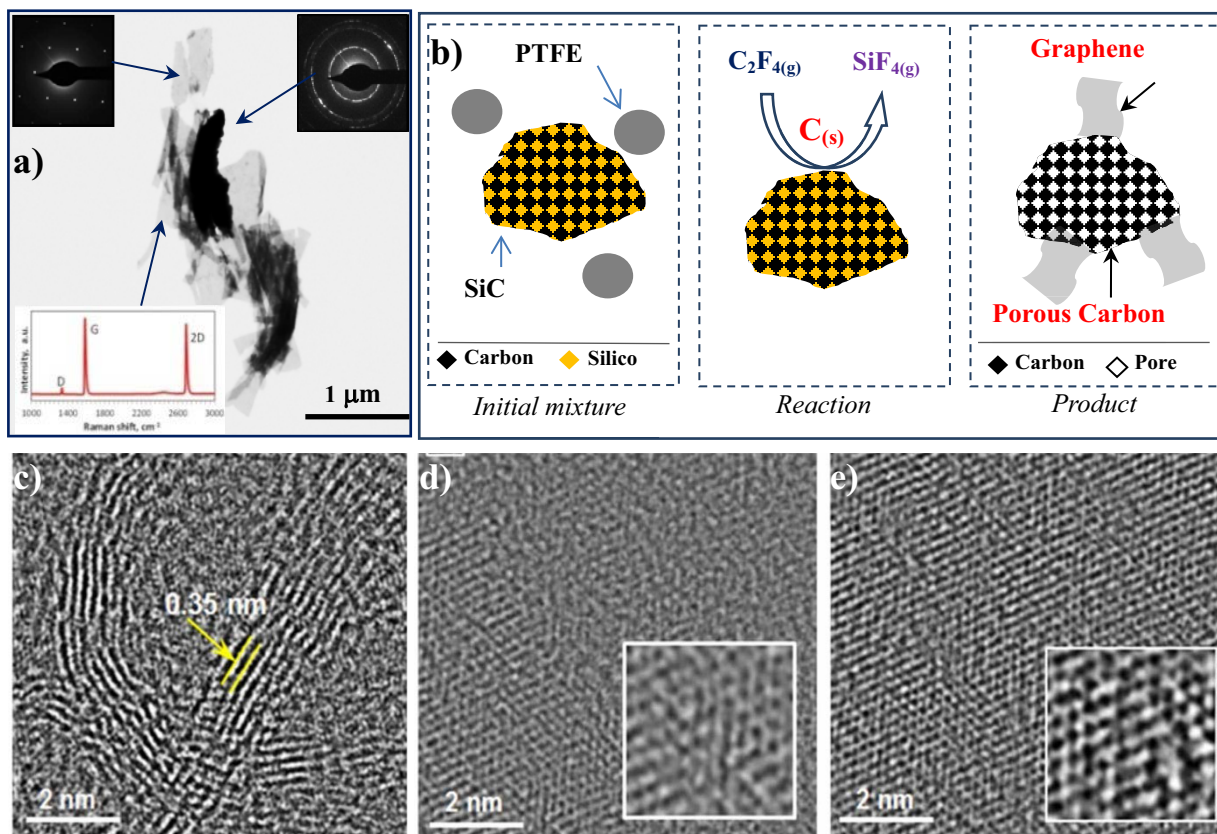


Figure 1 *Graphene – porous carbon* nano-structures (a), their synthesis scheme (b) and high resolution images of folded graphene layers on the surface of porous particles (c) and graphene flakes (d,e)

References

1. Novoselov, K. S., *et al.* Electric field effect in atomically thin carbon films. *Science* **306**, 666-669 (2004).
2. Geim, A. K., & Novoselov, K. S. The rise of graphene. *Nature Mater.* **6**, 183-191 (2007).
3. Stankovich, S., *et al.* Graphene-based composite materials. *Nature* **442**, 282-286 (2006)