The Mechanism of Catalytic Cutting of Graphene and Carbon Nanotube

Jinlan Wang¹, Liang Ma¹, Feng Ding²

¹ Department of Physics, Southeast University, Nanjing 211189, China
² Institute of Textiles and Clothing, Hong Kong Polytechnic University, Kowloon, Hong Kong, China

Email: jlwang@seu.edu.cn

Abstract

Synthesizing graphene nanoribbons (GNRs) with smooth edges and controllable widths is a prerequisite for many graphene applications.[1] By employing ab initio calculations, we proposed two efficient methods of making narrow GNRs: i) Using a single transition metal atom as the chemical scissor to unzip single-walled carbon nanotubes (SWNTs) into GNR in H2 gas;[2] ii) applying an external uniaxial strain to cut graphene sheet into GNRs upon oxidation.[3] The transition metal atom can drastically reduce the energy barrier of unzipping SWNTs, showing the feasibility of synthesizing high-quality narrow GNRs at only slightly elevated temperatures (~400K) (see Figure 1). The applied strain not only guides O atoms aligning along a zigzag direction that is closely perpendicular to the strain, but also significantly lowers the reaction barrier and the enthalpy of reaction of graphene cutting along that direction (see Figure 2).

Furthermore, by carefully modeling the TM particle-graphene interface and considering the role of hydrogen and oxygen in graphene edge termination, we successfully explain a long-term puzzle--why catalytic cutting of graphene by transition metal Ni, Co, and Fe nanoparticles is mostly along an armchair or zigzag direction (see Figure 3)? We also predicted that proper operating temperature and H2/O2 gas pressure can be used to tune the cutting behavior and thus allow the rationally experimental design for desired graphene structures [4].

References


Figures

Figure 1. a) Unzipping of an SWNT with a H2 molecule, b) Cu atom catalyzed H-H bond breaking, and c) Cu atom catalyzed C-C bond breaking on the edge of short (5, 5) SWNT. d) The feature cover picture of angew.chem.Int.Ed. The black, green, and pink balls represent carbon, hydrogen, and copper atoms, respectively.
Figure 2. a) Prefect graphene sheet. b)→c)→d) Random cutting graphene into quantum dots by oxygen attacking. e)→f)→g) orientation-selective cutting of strained graphene into GNRs by oxygen attacking.

Figure 3. (a) Catalytic cutting of graphene by metal nanoparticles along the armchair (left) and zigzag (right) edged channels. (b)→(e) The graphene-Ni(111) interfaces where the edges of the graphene are ZZ, (9,2), (2,1) and AC, respectively. Green, black and ultramarine balls represent H, C and Ni atoms, respectively. The interfacial carbon atoms are highlighted in red. (f) The graphene-Ni(111), Co(111), Cu(111) and Ag(111) interfacial formation energies as a function of the chiral angle of the graphene edge.