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Effect of hydrogen addition and temperature on morphology of thermal Ar/C_2H_2 plasma synthesized carbon nanomaterials

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Abstract:

Graphene nanoflakes accompanied with large accounts of carbon nanospheres are synthesized by thermal decomposition of acetylene using magnetically rotating arc at atmospheric pressure. The schematic diagram of the experimental setup is shown in Fig.1. According to observations of transmission electron microscopy (TEM) and high-resolution transmission electron microscopy (HRTEM), the morphologies and structures of carbon nanomaterials are analyzed. Tail gas composition and it concentration are investigated utilizing Gas chromatography. Firstly, low flow rate of hydrogen is added to the gas feed. It increases the relative graphene nanoflakes content of product. And near-pure graphene nanoflakes, which consist 2-10 sheets per stack with dimensions between 50 and 300 nm, can be obtained at higher H/C mole ratio (>8) of the gas feed mixture. It reveals that hydrogen inhibits acetylene conversion and thus reduces solid products. Subsequently, for the control group where the H/C mole ratio holds constant at 4, various the plasma arc power is investigated. As plasma arc power raising, products are almost graphene nanoflakes rather than carbon nanospheres. Experimental results indicate that hydrogen addition and reaction temperature increase bring about the morphological transformation of nanomaterials from carbon nanospheres to graphene nanoflakes.

The gas-phase kinetic model including the route to benzene and phenyl formation, PAHs growth from benzene to C20 is applied to investigate the role of hydrogen and temperature in reaction. The polycyclic aromatic hydrocarbons (PAHs) which are treated as a single graphene layer with hydrogen edge-passivated, is assumed as precursors to carbon nanospheres and graphene nanoflakes. Results of simulation demonstrates that hydrogen addition causes less efficient of hydrogen abstraction and acetylene addition (HACA) mechanism. Meanwhile, the rate of reverse reaction of PAHs formation is accelerative at the high temperature atmosphere. Hence, PAHs formation is inhibited by hydrogen addition and reaction temperature increase.

As is well-known, the morphology of carbon nanomaterials is determined by nucleation, collision and surface growth. Low concentration of large PAHs reduce the probability of collisions between them to induce two-dimensional nuclei formation due to stacking of fewer layers of graphene. It can be speculated that the slow rate of nucleation and rapid surface growth are beneficial for graphene synthesis. Thus, the key factor inducing the discrepancies in morphology of carbon nanomaterials under different H/C mole ratio of feed gas and temperature can be assigned to nucleation. Besides, hydrogen can terminate dangling bonds at edge and presumably maintain growing planar graphene sheets instead of fullerene-like shells. And five-member-ring is unstable at high temperature, making the growth process without curvature.

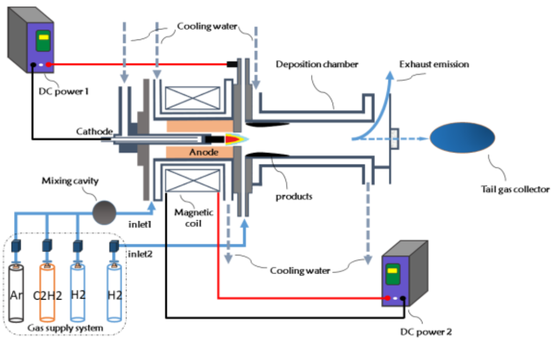

Fig. 1. Schematic diagram of the experimental setup