Low temperature thermal-CVD of methane

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Catalytic Vapour Decomposition (CVD) process is widely used for carbon nanostructures growth (CNT, Fullerenes, Graphene and etc.). The type of feedstock gas and temperature of synthesis are main parameters influencing efficiency of CVD. One possible way for decreasing synthesis temperature is use complex mixture of feedstock gases that have different decomposition temperature. Authors in [1] have shown that addition of acetylene or ethylene in methane leads to methane decomposition at low temperature about 500°C. Explanations of this temperature decrease are coadsorbtion of methane and acetylene/ethylene on surface of catalytic particles or phase transitions in catalytic particles due to exothermic reaction of acetylene/ethylene decomposition.

Experiments were carried out at thermal-CVD setup in conditions appropriate for CNT growth. A carbon containing gas mixture ($Ar/H_2+CH_4+C_2H_2/C_2H_4$) was supplied to quarts tube reactor after pre-warming with neutral gas flow. Iron catalySt was delivered to reactor hot zone by bubbling controllable portion of buffer gas flow through liquid iron pentacarbonyl at 0°C. Gas composition at the end of reactor was in situ controlled by gas analyzer RGA 200. Structures formed in reactor volume were collected at Teflon filter. Analysis of carbon soot was performed using TEM and SEM technique.

We observed that addition of small amount of acetylene/ethylene decrease temperature of decomposition methane however amount of decomposed methane equal to addition concentration. Therefore the most probable mechanism is formation of aromatics on surface of catalytic particles. Carbon soot collected from filter consists of CNT, onion like nanoparticles and amorphous carbon.

[1] Weizhong Qian,Tao Tian, Chenyi Guo, Qian Wen, Kuangjie Li, Hongbo Zhang, Haibo Shi, Dezheng Wang, Yi Liu, Qiang Zhang, Yongxiong Zhang, Fei Wei, Zhanwen Wang, Xiaodong Li, Yongdan Li. *J. Phys. Chem. C* **112**, 7588 (2008).