Catalytic Conversion of Graphene into Carbon Nanotubes via Gold Nanoclusters at Low Temperatures

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Introduction

 \succ Noble metal nanoparticles have huge potential in sensing, energy, spectroscopy, and catalysis.

 \succ Gold nanoclusters deposited on a variety of metal oxide support materials can efficiently catalyze oxidation and reduction reactions as well as C-C bond formation.

 \succ Au-based catalysts have been used in a range of organic transformations such as nucleophilic addition, cascade reactions, Fridel-Crafts reaction, C-H bond activation, hydrogenation and dehydrogenation reactions, asymmetric catalysis and total organic synthesis.

 \succ Synthesizing carbon nanomaterials on an Au catalyst or decorating graphitic nanostructures with AuNPs opens the door to a wide range of new applications.

 \succ Gold has been previously found to be a challenging catalytic element in the synthesis of CNTs and other carbon nanostructures.

> They are the first one to report the catalytic conversion of Au-decorated graphene to nanotubes at relatively low temperatures (500 °C) without the need of a hydrocarbon source by utilizing a radio frequency chemical vapor deposition (RF-CVD) reactor.

Experimental section



RF-CVD reactions were carried out under an Ar flow and without the addition of any hydrocarbon source, utilizing the functionalized graphene decorated with Au nanoclusters as the catalytic system.



SEM & TEM images of graphene sheets.

SEM & TEM images of Au decorated graphene sheets.

EDS maps confirms the presence of Au on the graphene sheets.



SEM images of the CNTs synthesized over the Au-decorated graphene sheets at 500 °C.

SEM images indicating high yield toward the synthesis of random networks of CNTs at 600 °C.

HRTEM images of the CNTs synthesized at 600 °C over the graphene sheets decorated with AuNPs.

SEM and TEM images at 650 °C. The SEM image indicates the presence of tangled tubular structures over the surface of graphene.



SEM and TEM images of the nanotubes synthesized on the Audecorated graphene sheets at 600 C.

SEM/EDS analysis indicating the presence of C, O2, and Au.



(b)

50 nm



SEM images of the synthesized CNTs on the Au-decorated graphene at 650 °C using ethylene.

TEM images of the synthesized CNTs on the Au-decorated graphene at 650 °C using ethylene.

TEM images showing tip growth mechanism.

This process was explained by two mechanisms:

- (1) In the absence of carbon source gas, under the thermal excitation, the AuNPs create small defects in the graphitic structure of the graphene layers and through a catalytic reaction process initiate the carbon nanotube formation. During the reaction, graphene decomposes providing a necessary source of carbon for further nanotube growth on the AuNPs.
- (2) In the presence of a carbon source, the catalytic nanotube growth is further enhanced due to the presence of a higher concentration of the carbon atoms. It may be possible that under these particular reaction conditions, graphene interacts with the AuNPs leading to the formation of nanotubes.



This figure presents the (TGA) mass loss curve of the commercial graphene and the Au-decorated graphene heated to 600 °C under Ar atmosphere.



RF reactions were performed on the nonfunctionalized graphene with and without acetylene at 600 and 650 °C. It is seen, after thorough SEM analyses that no CNT growth was detected at various temperatures.

Summary and conclusion

> AuNPs have the ability to convert the 2D structure of graphene into the 1D morphology of nanotubes, by the formation of C-C bonds that are required for the change in the crystalline geometrical structures of the two nanomaterials.

 \succ Non functionalized graphene sheets containing no nanoparticles or other impurities failed to yield CNT or other carbon nanostructure growth.

 \succ Graphene structures play a major role in the catalytic activity of AuNPs and that they can be responsible for the catalytic formation of CNTs.

> The catalytic reaction temperature (500 °C) was found to be lower than any other similar value reported in the literature, indicating that graphene could lower the CNT synthesis temperature, when used as a catalytic support material.

