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Synthesis of carbon nano-structured materials

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ABSTRACT

The deposition of carbon during catalytic reactions has a long history, with major efforts initially focused towards their prevention rather than synthesis. However the discovery of fullerenes and later that of carbon nanotubes by Iijima, shifted scientific focus towards the synthesis, characterization and application of carbon deposits. This renewed interest in carbon based materials, has revealed a universe of extraordinarily shaped carbon materials (SCMs) in the nano and micro range, from tubes and helices to horns and most recently graphene. It has been noted that there exists a relationship between the morphology of the carbon material and its inherent properties, making them highly prized for numerous technological applications. However before these carbon materials can be effectively exploited control over their selective synthesis is necessary, a problem that has been solved with only limited success. As such, there still exists a need to develop synthetic strategies that would yield shaped carbon materials selectively. More importantly, it is essential that a better understanding of the growth factors that lead to differently SCMs is obtained. In this study we have highlighted the parametric conditions for optimum growth of carbon helices, as well as that of carbon fibers with unique structure. We have found that catalyst morphology and the carbon source are key aspects, which control carbon material growth and morphology.

The synthesis of carbon materials using bi and tri-metallic supported catalyst systems revealed that Cu was an effective promoter for obtaining helices, particularly at low temperatures (≤ 550 ˚C). On further investigation, Cu was shown to exhibit incredible carbon deposition capabilities at temperatures as low as 200 ˚C. Adjustments of the catalyst preparation conditions (support, metal counter ion, solvent and reduction temperature) and synthesis temperature, revealed that the yield and morphology of the carbon deposit could be altered to selectively produce both straight and helical carbon fibers.

A TEM tomography study revealed that the copper particles that gave distorted decahedra formed helical fibers, while trigonal bi-pyramidal particles gave linear fibers. Various plate-like particles revealed that as the number of sides of a catalyst particle varied (3, 4, 5 or 6) there was a corresponding change in the

carbon fiber helicity. A relationship between catalyst particle morphology and fiber morphology was thus established.

TEM analysis also revealed that catalyst particles underwent rapid reconstruction during carbon fiber synthesis, and that the carbon source (gas environment) was influential in this reconstruction event. A NiO_x (unsupported) catalyst was prepared and reactions with various substituted alkyne hydrocarbons were undertaken. Analysis revealed that different alkynes produced carbon fibers with varying morphologies. Using different alkynes in a sequential manner led to the formation of 'co-block' carbon fibers with an A-B-A-B... or A-B-C... morphology. Using different alkynes followed by acetylene led to the selective synthesis of straight, Y-junction or irregular carbon fibers. Accompanying these results was the observation that in each case the catalyst particle morphology was unique. Reaction of NiO_x with trichloroethylene, in which trichloroethylene acted as a source of carbon for fiber growth, also restructured the Ni catalyst into a tetrahedral shape that gave tripod-like carbon growth. It was found that, substituted alkynes (and alkenes) provided a means for controlling catalyst particle morphology and hence carbon fiber morphology.

The study has highlighted the relationship that exists between catalyst and SCM morphology, as well as the effect of hydrocarbons, not only as a source of carbon for SCM growth but also as a means of controlling catalyst morphology and SCM structures.