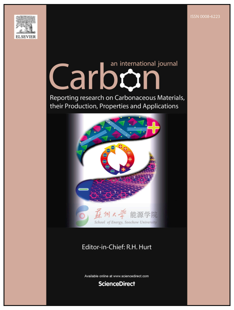

The Influence of Magnetic Moment on Carbon Nanotube Nucleation

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The Influence of Magnetic Moment on Carbon Nanotube Nucleation

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Abstract

We demonstrate the impact of magnetic moment on single-walled carbon nanotube (SWCNT) nucleation, using quantum chemical molecular dynamics simulations. The mechanism of SWCNT cap formation on Fe_{38} nanoparticle catalysts can be manipulated by altering the total magnetic moment. When the magnetic moment is low, large cap structures dominated by pentagons are formed. Increasing the magnetic moment leads to smaller, "flatter" sp^2 carbon networks with a higher proportion of hexagons, which are incapable of lifting away from the caõtalyst surface. These results demonstrate that SWCNT (n,m) chirality distributions can be potentially manipulated via altering the magnetic moment of the catalyst-carbon interface. We also show that higher magnetic moments leads to slower SWCNT nucleation. This is a consequence of the extent of Fe→C charge transfer, which is proportional to the total magnetic moment. As the magnetic moment is increased, the Fe_{38} nanoparticle has higher catalytic activity and is able to stabilise carbon in its subsurface region, thereby impeding the nucleation process.

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1. Introduction

Single walled carbon nanotubes (SWCNTs) possess remarkable physicochemical, electronic and mechanical properties [1–4], which give them great potential in diverse technological applications [5]. The electronic and optical properties of a SWCNT are dependent on its (n,m) chiral indices. Since their initial characterisation [6, 7], traditional SWCNT synthesis methods, such as the arc discharge and chemical vapor deposition (CVD), have struggled to produce *in situ* (n,m) chirality SWCNTs selectively (so-called "chirality controlled growth"). This problem has represented the single largest hurdle in producing new SWCNT-based technologies over the last 20 years.

CVD growth of CNTs is currently believed to be the most promising approach for producing larger quantities of CNTs with controlled chiralities, since it enables more direct control over the physicochemical parameters that govern the nucleation/growth mechanism. The most widely used catalysts in this respect have traditionally been iron-group metals (Fe, Co and Ni) [8]. Recent advances based on catalyst design approaches (*i.e.* by tailoring the catalyst's elemental composition, phase and surface morphology) have enabled exclusive/near-exclusive growth of single (n,m) SWCNTs [9, 10]. Similar advances have also been made with cloning/vapor-phase epitaxy approaches [11–14], and the use of organic templates for SWCNT growth (*e.g.* cycloparaphenylenes) [15, 16].

At the molecular level, CNT nucleation and growth is an immensely complex process controlled by experimental parameters including temperature [17], catalyst type [18–23], catalyst particle size [24, 25], the type of carbon feed stock and feedstock pressure [19, 26, 27], the presence of reducing agents [28] (notably hydrogen [23, 29–34]), and applied

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