

Alignment Control of Carbon Nanotube Forests from  
Random to Nearly Perfectly Aligned by Utilizing  
Crowding Effect

Ming Xu<sup>1,2,3</sup>, Don N. Futaba<sup>1,2</sup>, Motoo Yumura<sup>1,2</sup>, Kenji  
Hata<sup>1,2,4</sup>

<sup>1</sup>Technology Research Association for Single Wall  
Carbon Nanotubes (TASC), Tsukuba, 305-8565, Japan

<sup>2</sup>Nanotube Research Center, National Institute of  
Advanced Industrial Science and Technology (AIST),  
Tsukuba, 305-8565, Japan

<sup>3</sup>College of Materials Science and Engineering, Huazhong  
University of Science and Technology, Wuhan, 430074,  
P. R. China.

<sup>4</sup>Japan Science and Technology Agency (JST),  
Kawaguchi, 332-0012, Japan

As in the case for polymer chains, fibers, and proteins, alignment of the one-dimensional, high aspect ratio internal building blocks of a bulk material represents an important structure parameter. Due to the impact of alignment on properties, tailoring the degree of alignment of these components is of fundamental importance. An apt example is polyethylene (PE) used to make drink bottles where the hard cap is made from highly ordered PE while the flexible body is made from low order PE.

As being a one-dimensional material possessing the highest aspect ratio, carbon nanotubes (CNTs) follow a similar scheme. In this paper, we demonstrate a general approach to control the alignment of few-walled CNT forests from nearly random to nearly ideally aligned by tailoring the density of active catalysts at the catalyst formation stage, which can be experimentally achieved by controlling the CNT forest mass density. We found that the catalyst density and the degree of alignment were inseparably linked because of a crowding effect from neighboring CNTs, i.e., the increasing confinement of CNTs with increased density. Therefore, the CNT density governed the degree of alignment, which increased monotonically with the density. This relationship, in turn, allowed the precise control of the alignment through control of the mass density. The wide variety of CNT forests with different alignments achieved in this work would be expected to open new opportunities for different CNT structures and forms or new post-processes.