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Shape Control of Carbon Nanotube Forests via Bottom-up Process of Catalyst Nanoparticles

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High-areal-density, vertically aligned carbon nanotubes (VACNTs), commonly referred to as carbon nanotube forests, have been widely studied for their unique structural and functional properties. Prior research has demonstrated their applicability in diverse fields such as field emission, thermal interface materials, and energy harvesting, with particular emphasis on achieving dense and vertically aligned morphologies on various substrate surfaces. Because CNT forests are typically synthesized through a base-growth mechanism from nanoscale catalyst particles placed on substrates, various efforts have been made to produce forests with smaller diameter, higher-areal-density, and increased height. Catalyst modification processes are among the most widely employed strategies to form high-areal-density, small-diameter catalyst seeds on substrates, which are essential for initiating and sustaining CNT growth.

For instance, J. Robertson reported that plasma treatment on Al2O3 buffer layers enabled the formation of CNT forests with densities as high as 1.5×10^{13} CNTs/cm^2 [1]. Maruyama et al. demonstrated the anchoring effect of bimetallic catalysts [2], such as Co-Cu, which improved catalyst stability and promoted vertically aligned growth. We have also reported an interval RF sputtering method to produce small-diameter catalyst particles suitable for single-walled carbon nanotube (SWNT) growth, where higher-density plasma enhances the anchoring effect of iron catalysts onto the Al_2O_3 buffer layer. Additionally, we demonstrated that oxidizing the Al_2O_3 buffer layer prior to Fe catalyst film deposition increases the areal density of catalyst particles after annealing. These results support the conclusion that the interfacial anchoring effect is crucial for producing CNT forests with high areal density and small core diameters.

Advanced techniques for shape control of CNT forests through the annealing of multi-layered catalyst thin films deposited by magnetron sputtering will be discussed in this paper. The transformation of these films into nanoparticles involves high temperature annealing in reducing atmospheres. A key aspect, investigated using mass spectrometry, was the effect of the furnace environment, specifically residual gases like water vapor (H_2O) and oxygen (O_2), during this catalyst formation stage (Fig.1) [3, 4]. We found residual H2O promoted CNT elongation, while O2 appeared to suppress catalyst aggregation, influencing density [4]. Furthermore, specific gas desorption patterns correlated with effective nanoparticle formation and longer CNT growth [4].

Controlling these annealing conditions (including atmosphere) alongside the multi-layer structure enables precise tuning of catalyst nanoparticle size and distribution, significantly influencing VACNT growth behavior and morphology (Fig.2). By leveraging nanoscale engineering approaches, we demonstrate tunable CNT forest architectures with high reproducibility. These structures exhibit promising characteristics for next-generation applications, including neuromorphic electronics, solar thermal energy storage, and advanced thermal management systems.

References
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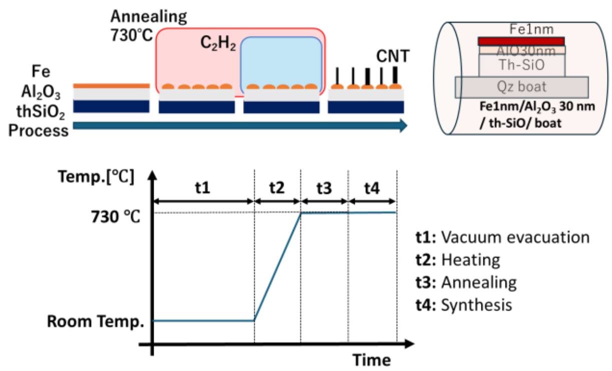

Figure 1 Annealing process of AlO/Fe multilayered catalyst films with control H_2O/O_2 residual gas in vacuum chamber[3,4].

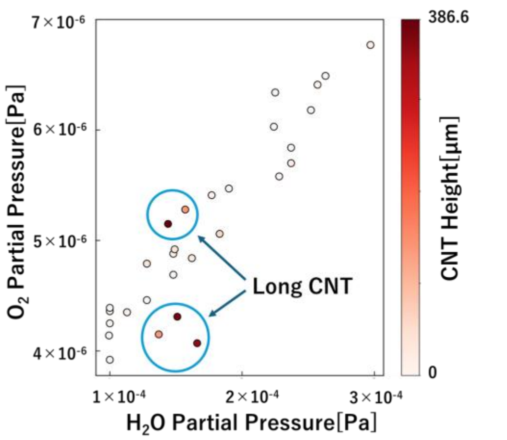

Figure 2 Growth height of CNT forests with various H_2O/O_2 residual partial pressure in the annealing process[4].