# Temperature Dependence of Carbon Nanotube Growths

**Joshua Alden**

**Biomedical Engineering, Georgia Institute of Technology**

***NNIN REU Site: Cornell NanoScale Science and Technology Facility, Cornell University, Ithaca, NY***

*NNIN REU Principal Investigator: Paul McEuen, Department of Physics, Laboratory of Atomic and Solid State Physics and Kavli Institute for Nanoscale Science, Cornell University*

*NNIN REU Mentor: Alejandro Cortese, Department of Physics, Cornell University*

*Contact: joshualden@gmail.com, pmceuen@gmail.com, ajc383@cornell.edu*

## Introduction:

Carbon nanotubes (CNTs) have unique electrical and physical properties that make them candidates for many different applications ranging from high-speed field-effect transistors to biosensors. For reliability and efficiency in such applications, it is important to run many devices in parallel, which requires nanotubes to be grown in consistent arrays at appropriate densities. In this project we explored how nanotube growth temperature is correlated with carbon nanotube density, nanotube diameter, and catalyst size. We also investigated the accuracy with which an atomic force microscope (AFM) measures nanotube diameter. Being nature's smallest wire, nanotubes have the potential to act as biosensors capable of sensing small molecules such as individual nucleotides in DNA. The sensitivity of these biosensors would then be dependent on the nanotube diameter, thus making it an important point of study.

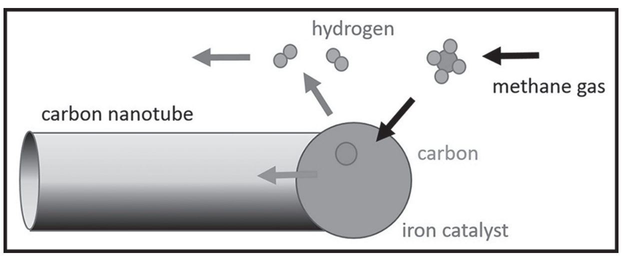


*Figure 1: Chemical vapor deposition. Methane gas flows near an iron catalyst which absorbs the carbon into itself, and after saturation, the carbon moves to the outside of the catalyst and begins to form a nanotube.*

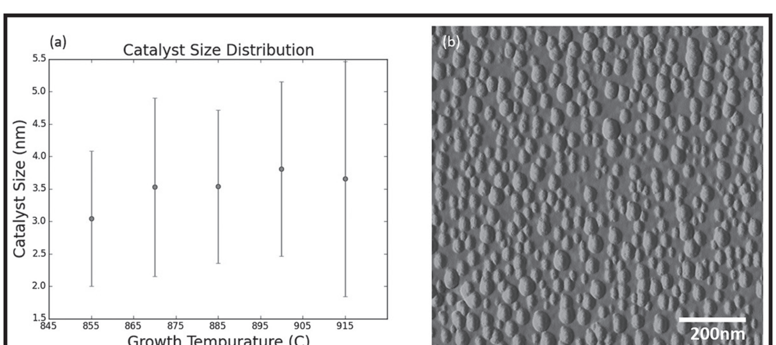


*Figure 2: a) Catalyst size increased with increasing temperature. The dots and lines show respectively, the mean and standard deviation of observed catalyst sizes by AFM. b) AFM image of catalysts after an 870°C growth.*

## Experimental Procedure:

Carbon nanotubes are grown from iron catalyst nanoparticles on quartz wafers in a high temperature chemical vapor deposition process, as illustrated in Figure 1. The quartz wafers used are ST cut, which means that they have atomic-scale terraces running in parallel across the wafer. Nanotubes can grow along these, leading to arrays of aligned nanotubes. Wafers were fabricated and nanotubes grown using standard procedures [1,2].

Following growth, AFM imaging was used to collect the heights and thus the diameters of the iron catalyst particles after growth and the diameters of the nanotubes while SEM imaging was used to measure the density of nanotubes after each growth. To calibrate the AFM nanotube diameter measurements, AFM-measured nanotubes from a quartz substrate were transferred onto TEM grids. The nanotubes were then imaged again using transmission electron microscopy (TEM) and their diameters were measured.

## Results and Discussion:

In the temperature range from 855°C to 915°C, we observed that average catalyst diameter increases with increasing temperature as shown in Figure 2a. However, the width of the distribution of catalyst sizes within each growth was larger than the observed increase. Ostwald ripening is the phenomenon suspected to be causing this trend. As temperature goes up, the iron atoms gain the energy necessary to migrate from smaller catalyst particles to larger ones, since this minimizes the number of atoms exposed at the surface. As a result, larger catalysts form during higher temperature growths.

We also observed a strong temperature-dependence to number of nanotubes per micron that grew from a given catalyst line, as shown in Figure 3. We noticed that CNTs grown at 855°C have low number density as can be seen in Figure 3a. Between 870°C and 885°C, the nanotubes grow at high densities, as in Figure 3b. Above 885°C, the density tapers off to 0, as seen in the image in Figure 3c. Because nanotubes grow from the surface of catalyst particles, catalyst size and nanotube density are linked together. Once the catalysts reach an optimal size, many nanotubes will grow thus creating a high density of CNTs at that temperature. But at higher temperature growths, many of the catalysts are too large to initiate growth as resulting in a lower density as observed in Figure 3d.

Figure 4a shows AFM measurements of the average nanotube diameter as a function of temperature. The average nanotube diameter increases with increasing temperature. To calibrate these measurements, which can have systematic error due to tip-substrate interactions, one of the samples was AFM'd, then TEM'd. The resulting histogram of observed diameters is shown in Figure 4b. The more-accurate TEM measurements measured larger nanotube diameters than the AFM (Figure 4b).

In summary, we found that catalyst size, and nanotube diameter increase with increasing temperature and that nanotube density is tightly linked with the catalyst size. Knowing the temperature-dependence of carbon nanotubes on temperature allows us to reliably grow nanotubes of a pre-selected density and diameter, bringing wafer-scale production of nanotube biosensor arrays one step closer to viability.

## Future Work:

Having developed an understanding and methodology for nanotube growth in a 5-inch furnace, we can now begin testing 4-inch wafer growths. If our methods successfully scale, this work will be used in the McEuen group's work on developing arrays of DNA sequencing devices. Other interesting expansions of this project include varying parameters such as the anneal time, and hydrogen and methane flow times and rates to create more uniform catalyst sizes with a narrower distribution. This may allow for more precise control over nanotube diameter and possibly a more exact density-temperature relationship.


## Acknowledgments:

I would like to thank my mentor Alejandro Cortese and fellow lab members Jonathan Alden and Bryce Kobrin for their help and support on this project. I would also like to thank the McEuen lab and CNF for a positive and productive work environment. This research was supported by the National Nanotechnology Infrastructure Network Research Experience for Undergraduates (NNIN REU) Program and National Science Foundation under Grant No. ECCS-0335765.


## References:


[1] Ding, L.; Yuan, D.; Liu, J. "Growth of High-Density Parallel Arrays of Long Single-Walled Carbon Nanotubes on Quartz Substrates." J. Am. Chem. Soc. 130, 5428.5429, 2008.

[2] Kocabas, C.; Hur, S.-H.; Gaur, A.; Meitl, M. A.; Shim, M.; Rogers, J. A. "Guided Growth of Large-Scale, Horizontally Aligned Arrays of Single-Walled Carbon Nanotubes and Their Use in Thin Film Transistors." Small 1, 1110.1116, 2005.


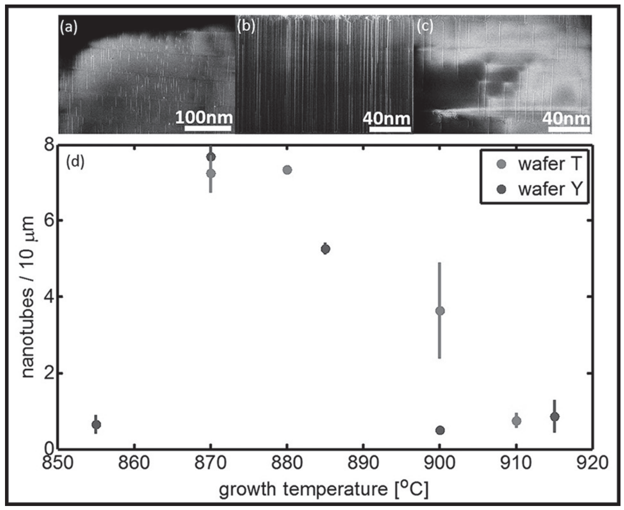


*Figure 3: a) SEM image of nanotubes after an 855°C growth. b) SEM image of nanotubes after an 885°C growth. c) SEM image of nanotubes after a 915°C growth. d) Nanotube Density vs. Temperature. The dots and lines show respectively, the mean and standard deviation of the measured nanotube densities. The two different wafers are from two different fabrication batches and growths.*

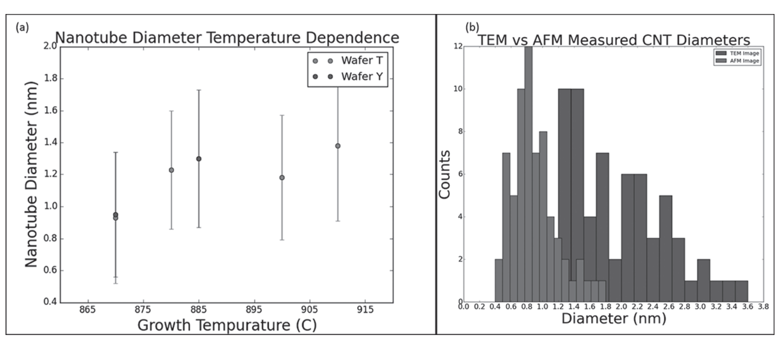


*Figure 4: a) Shows the mean and standard deviation of AFM measured nanotubes from two separately fabricated wafers. b) Shows a histogram of the nanotube diameters measured by the AFM and TEM.*