Anisotropic thermal conductivity of aligned carbon nanotube arrays

T. Borca-Tasciuc  
Department of Mechanical, Aerospace, and Nuclear Engineering, Rensselaer Polytechnic Institute, Troy, NY, USA

R. Vajtai, B. Q. Wei, & P. M. Ajayan  
Department of Materials Science and Engineering, Rensselaer Polytechnic Institute, Troy, NY, USA

M. S. Shur  
Electrical, Computing, and Systems Engineering Department, Rensselaer Polytechnic Institute, Troy, NY, USA

J. Deng & R. Gaska  
Sensor Electronic Technology, Inc., Latham, NY, USA

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**ABSTRACT:** By improving the thermal conductivity of both the substrates and the package materials, carbon nanotube technology is expected to address the self-heating problem, which seriously degrades the device performance in high-power electronic and optoelectronic devices. In this work we present our ongoing efforts in the fabrication and thermal properties characterization of structures with expected high thermal conductivities based on aligned carbon nanotube arrays. The arrays are grown on selective sites on silicon substrates and into self-organized anodic aluminum oxide films with deep submicron periodic honeycomb structures. The anisotropic thermal conductivity measurement of the carbon nanotube filled anodic alumina samples is based on a differential 2-wire $3\omega$ method. Heaters and temperature sensors instrumented onto patterned mesa structures are employed for the thermal characterization of the aligned carbon nanotube films grown on silicon substrates.

1 **INTRODUCTION**

The controlled growth of high purity, aligned, carbon nanotube (CNT) arrays (Zhang et al. 2000) paves the way for the possible use of CNTs as field emission sources, interconnects for 2D and 3D IC architectures, and heat transport media for the thermal management of electronics, optoelectronics, and solid state cooling devices. CNTs structures with large thermal conductivities are highly desirable, and theoretical predictions indicate that the thermal conductivity along the length of a single CNT may be equal or even larger than the thermal conductivity of diamond or graphite (Berber et al. 2000, Osman & Srivastava 2001). However, reported values of the experimentally measured thermal conductivities for several carbon nanotube structures are typically much lower than the theoretical predictions (Hone et al. 1999, Yi et al. 1999, Hone et al. 2000). Growth defects, entanglements of the carbon nanotubes ropes, and imperfect alignment, have been proposed as possible causes for the observed thermal conductivity reduction. Experimental measurements of the thermal conductivity in CNT structures have been reported for high-purity “mats” of tangled nanotube bundles (Hone et al. 1999), millimeter long bundles of aligned multiwall CNTs (Yi et al. 1999), and magnetically aligned single wall CNTs films (Hone et al. 2000). Removal of the CNTs from the deposition substrate was required to perform the alignment of the nanotubes (Hone et al. 2000) or the thermophysical characterization (Yi et al. 1999). Furthermore, the previous experimental reports are focused on thermal characterization along, or in the plane, of the alignment direction. However, the anisotropic configuration of the aligned CNT structures is expected to induce
a highly anisotropic thermal conductivity, which could be important in the thermal management applications of CNT structures.

In this work we present our ongoing efforts in the fabrication and characterization of the anisotropic thermal properties of high purity, aligned carbon nanotube arrays grown on selective sites on silicon substrates and into self-organized anodic aluminum oxide films with deep submicron periodic honeycomb structures. A differential 2-wire 3ω method is proposed for the anisotropic thermal conductivity measurement of the carbon nanotube filled anodic alumina samples. Heaters and temperature sensors instrumented onto patterned mesa structures are employed for the thermal characterization of the aligned carbon nanotube films grown on silicon substrates.

2 SAMPLES AND MEASUREMENT TECHNIQUE

Figure 1 shows a SEM micrograph of an aligned CNT film selectively grown over a 100nm SiO₂ film deposited onto the silicon substrate. The CNT bundles grow perpendicular to the substrate and are self-standing. The total thickness of the CNT film is ~ 90µm. A thin layer of amorphous carbon is present on the top surface of the film. A more mechanically stable structure could be obtained by incorporating carbon nanotubes into self-organized aluminum anodic oxide films with deep submicron periodic honeycomb structures. Figure 2 shows a SEM picture of an anodic alumina sample, which serve as a template for CNT growth. The individual pores are ~40nm in diameter. The aluminum oxide with carbon nanotubes will be an alternative substrate, which may have superior thermal performance to SiC. For example assuming the voids are filled with carbon nanotubes with a thermal conductivity of 30 W/K-cm along the tube and a 40% initial porosity of the template (the ratio of the void area compared to the whole area), the thermal conductivity of CNT filled alumina substrates can be as high as 12W/cm-K, two times higher than SiC substrate. By improving the thermal conductivity of both the substrates and the package materials, carbon nanotube technology is expected to address the self-heating problem, which seriously degrades the device performance in high-power electronic and optoelectronic devices. Figure 3 shows a proposed example for using such arrays for improved heat sinking of high power GaN-based transistors.

The anisotropic thermal conductivity measurement of the carbon nanotube filled anodic alumina samples is based on a differential 2-wire 3ω method (Borca-Tasciuc et al. 1998, Borca-Tasciuc et al. 2000). A similar method was used by Kumar et al. (2000) for the thermal conductivity charac-
Figure 2. SEM micrograph of the top surface of a self organized aluminum anodic oxide films with deep submicron periodic honeycomb structures, which is used as a template for CNT growth.

Figure 3. Possible heat sinking scheme for high power AlGaN/GaN HFETs using high thermal conductivity alumina templates with carbon nanotubes.

Figure 4. Anisotropic thermal conductivity characterization using microfabricated heaters/temperature sensors with different widths deposited on the alumina templates and carbon nanotube filled samples.

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The anisotropic thermophysical properties of the samples are determined by fitting the experimental temperature rise collected for different wire widths with predictions of heat transport models based on 2D heat conduction in a multilayer-film-on-finite-substrate structure (Borca-Tasciuc et al. 2001). Figure 5 shows the predicted thermal response of heaters with different widths deposited over both empty and CNT filled alumina templates. The simulations consider free-standing 75 \( \mu \text{m} \) thick alumina templates with porosity 35%, 40nm pore diameter, and assumes the channels are only partially filled (20%) with multi-wall CNT of thermal conductivity 1000W/mK. The effective thermal conductivity of the empty template was taken as 1W/mK along the direction of the channels and 0.7W/mK along the direction perpendicular to the channels. The contribution of the CNT to heat conduction in the direction perpendicular to the channels was neglected. As shown in Figure 5a, the temperature signal is more sensitive to the thermal transport along the channels as frequency increases and heater width decreases. At low frequencies and for large width heaters, the temperature signal is more sensitive to transport in the plane of the alumina template film. Figure 5b shows the phase of the thermal signal. The shift of the phase peak to larger modulation frequencies is due to the increase in thermal diffusivity along the channels of the simulated CNT filled template.

The same technique could be employed in principle to obtain the anisotropic thermal conductivity of the aligned CNT films shown in Figure 1. However, typical cleanroom microfabrication of metallic heaters and temperature sensors on the CNT film is difficult, partially due to the mechanical fragility of the films. Therefore, we used a different approach to pattern heaters and temperature sensors on the top surface of the film. Since the CNT films grow selectively over the SiO\(_2\) surface, we patterned “heaters” on the SiO\(_2\) film before the growth. The result after the growth of the CNT is shown in Figure 6. Millimeter long mesa structures with different heater widths and the same thickness as the CNT film are easily obtained. The amorphous carbon layer on the top surface could be used as a heater or a metallic film could be evaporated on the top surface. A differential 3\( \omega \) method is employed to measure the temperature difference between same width heaters on top of CNT “mesas” with different thickness. Figure 7 shows a schematic of the differential measurement technique used to determine the thermal conductivity along the CNT growth direction. After etching the SiO\(_2\) layer underneath the long “mesas”, the structures can be used to determine the thermal conductivity of the CNT film in the direction perpendicular to the CNT growth.
Figure 6. SEM micrograph of the top view of a sample with a selectively grown CNT film forming long “mesas” on the surface. The top surface of the “mesa” is electrically conductive due to the presence of a thin amorphous carbon layer. Evaporation of a thin metal layer could be performed to increase the electrical conductivity. The numbers indicate the width (in microns) of the wire between the inner contact pads.

Figure 7. “Mesas” of aligned CNT films with different thickness are employed in a differential 3ω technique to determine the thermal conductivity of the film along the direction of CNT growth. After etching the SiO$_2$ layer underneath the long “mesas”, the structures can be used to determine the thermal conductivity of the CNT film in the direction perpendicular to the CNT growth.

3 CONCLUSIONS

We present our efforts in the fabrication and thermal properties characterization of structures based on aligned carbon nanotube arrays with expected high thermal conductivities. CNT arrays are grown on selective sites on silicon substrates and into self-organized anodic aluminum oxide films with deep submicron periodic honeycomb structures. The differential 2-wire 3ω method will be used for the anisotropic thermal conductivity measurement of the carbon nanotube filled anodic alumina samples. Heaters and temperature sensors instrumented onto patterned mesa structures are proposed for the thermal characterization of the aligned carbon nanotube films grown on silicon substrates. Thermal conductivity characterization of aligned CNT arrays is currently under investigation.

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