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Thermal performance of vertically-aligned multi-walled carbon nanotube array grown on platinum film

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A B S T R A C T

Vertically-aligned carbon nanotube array is expected to inherit high thermal conductivity and mechanical compliance of individual carbon nanotube and serve as thermal interface material. In this paper, vertically-aligned multi-walled carbon nanotube arrays have been directly grown on Pt film and the thermal performance has been studied by using laser flash technique. The determined thermal diffusivity decreases from 0.187 to 0.135 $cm^2\,s^{-1}$ and the thermal conductivity increases from 1.8 to 3.1 $W\,m^{-1}\,K^{-1}$ as temperature increases from 243.2 to 453.2 K. The fracture surface of the array peeled off the Pt film was characterized by scanning electron microscopy. It has been illustrated that the tearing surface is not smooth but fluffy with torn carbon nanotubes, indicating strong interfacial bonding and consequent small interface resistance between carbon nanotube array and Pt film. According to Raman spectra and transmission electron microscopy image, the possible mechanisms responsible for the thermal transport degradation are low packing density, twist, and the presence of impurities, amorphous carbon, defects and flaws. The influence of intertube van der Waals interactions has been studied by comparing the phonon dispersion relations and is expected to be not significant.

1. Introduction

With rapidly increasing power densities in electronic devices, thermal management is becoming a crucial issue that limits the performance, power, reliability and further miniaturization of microelectronics [1]. Although heat spreaders and heat sinks of high thermal conductivities have been employed to dissipate the heat generated from the die, thermal resistances

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imposed by limited actual contact at multiple interfaces from the die through the heat spreader to the outside heat sink undermine the effective thermal conduction and remain a potential bottleneck. Thermal interface materials (TIMs) have been introduced to reduce or completely eliminate the microscopic cavities from the contact interfaces and consequently enhance the heat transfer by conforming to the rough and uneven mating surfaces [2,3].

Vertically aligned carbon nanotube (CNT) arrays, which consist of many flexible nanotubes aligned from one surface to the opposite side, may eventually provide both high thermal conductivity and lateral compliance, have been considered as a promising candidate for TIMs [4–15]. Significant enhancement of the overall thermal conductance can be expected, since all the CNTs are forming ideal thermal conducting paths from one surface to the other with extremely high intrinsic thermal conductivity of individual CNTs. Experimental and molecular dynamic studies have determined thermal conductivities of individual single-walled CNTs (SWCNTs) in the range of 2500–6600 W m^{-1} K^{-1} [16–18] and multi-walled CNTs (MWCNTs) in the range of 2000–3000 W m^{-1} K^{-1} [19,20] at room temperature. CNT array is expected to inherit the high thermal conductivity of individual CNT.

Characterization of the thermal performance of CNT array has been a recent research focus and a considerable amount of work [4–15,21–32] has been carried out. In general, these studies have reached a consensus on the following aspects: (1) The CNT array has a higher thermal conductivity than most regular TIMs applied in industry nowadays; and (2) the thermal conductivity of CNT arrays is much lower than that of single CNT. However, the determined thermal conductivities of CNT arrays scatter over a wide range, from 0.145 to over 200 W m^{-1} K^{-1} [5,7,8,14,22–24,26,27,29–32], as illustrated in Table 1. The first reason responsible for the scatterings is that the CNT arrays are intrinsically different. For instance, the thermal conductivity of the MWCNT array significantly increases after 2800 °C annealed, from 2.7 to 23.4 W m^{-1} K^{-1} [27]. Besides, different determination methods as well as assumptions may also induce the scatterings. As illustrated in Table 1, the assumed or determined density of the CNT arrays scatters from 11.72 to 1300 kg m^{-3}. Although the packing density of the CNT arrays are different, the difference in the density cannot be so large, which dominates the thermal conductivity via $\lambda = \alpha\rho C_p$, where α, ρ, and C_p denote the thermal diffusivity, density and specific heat, respectively. In addition, most of the previous studies usually focused on the thermal transport properties of the CNT arrays at room temperature. As TIM in application, the thermal transport properties above and below room temperature are also very important.

The thermal interface resistance at the CNT array/mating substrate interface is another key issue. It has been found that the thermal interface resistance is large and dominates the total thermal resistance of the CNT array TIM. Tong et al. [10] studied the TIM formed by 7-μm-thick MWCNT array grown on Si and directly dry adhered to glass. The determined total thermal resistance was 12 m^2 K MW^{-1} and dominated by the interface resistance between the free CNT array tips and the opposing glass of 11 m^2 K MW^{-1}, while the CVD growth CNT array/Si interface resistance is one order of magnitude lower. Cola et al. [13] also reported that the PECVD grown CNT array/Si interface resistance is only 1/7 of that of free CNT array tips/Ag foil. Panzer et al. [14] found poor contact between the SWCNT array and evaporation coated Pd and only 2.9% of the SWCNTs contributed to the thermal transport, which induced large effective thermal interface resistance. Direct growth of the CNT array on the contact surface is suggested as a promising approach for improving the thermal performance of CNT array-based interface materials [5,14,31]. Attention has been usually focused on growth of the CNT array on Si substrate. However, its practical application is still limited due to the temperature restriction of current electronics. Hence, growth of CNT array on pure metal and metal alloy is expected to be applied in direct growth of the CNT array on heat sink and fabrication of the two-sided CNT array TIM (Si–CNT–CNT–metal) [11,13,24,31].

In this paper, the thermal performance of two MWCNT arrays of different thickness grown on Pt film in the temperature range from 243.2 to 453.2 K has been studied by applying the laser flash method. The fracture surface of the MWCNT array peeled off the Pt film has been characterized by scanning electron microscopy (SEM). The SEM images show that the tearing surface is not smooth but fluffy with torn CNTs, indicating strong interfacial bonding and consequent small

Table 1 – Reported thermal conductivities of CNT arrays.

Author	CNT array	λ (W m^{-1} K^{-1})	Method
Hu et al. [5]	MWCNT	~75	3-omega method.
Shaikh et al. [8]	CNT	8.3	Laser flash method
Xu et al. [7]	CNT	2.85	Photothermal method, ρ = 185 kg m^{-3}
Panzer et al. [14]	SWCNT	8	Thermoreflectance method
Hone et al. [22]	SWCNT	~217	Comparative and self-heating method
Yang et al. [23]	MWCNT	12–17	Photothermal method
Wang et al. [24]	MWCNT	0.145	Photothermal method, ρ = 11.72 kg m^{-3}
Ivanov et al. [26]	MWCNT	3.0, 5.7, 6.4, 10, 15	Laser flash method, ρ = 153.1 kg m^{-3}
Jin et al. [27]	MWCNT	~2.7, 3.3, 8.1, 23.4	Physical property measurement system
Jakubinek et al. [29]	MWCNT	0.5–1.2	Steady-state technique
Lin et al. [30]	MWCNT	27	Laser flash method, ρ = 1300 kg m^{-3}
Sunil et al. [31]	MWCNT	0.8	Steady-state technique
Akoshima et al. [32]	SWCNT	1.9	Laser flash method, ρ = 40 kg m^{-3}

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