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A Meta-study on the Thermal Properties of Carbon Nanotubes in Thermal Interface Materials

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Abstract: Computer Integrated Circuit (IC) microprocessors are becoming more powerful and densely packed while cooling mechanisms are seeing an equivalent improvement to compensate. A significant limit to cooling performance is thermal transfer between die and heatsink. In this meta study we evaluate carbon nanotube (CNT) thermal interface materials (TIMs) in order to determine how to maximise thermal transfer efficiency. We gathered information from over 15 articles focused on the thermodynamic parameters of CNT TIMs from databases such as Scopus, IEEE Xplore and ScienceDirect. Articles were filtered by key words including 'carbon nanotubes' and 'thermal interface materials' to identify scientific articles relevant to our research on TIMs. From our meta study we have found that enhancing CNTs will provide the best improvement in TIMs. The parameters analysed to determine TIM performance included thermal resistance, thermal conductivity and the effect of CNT concentration on computer operation time. Through our investigation we understood that increasing the concentration of CNT from 0 to 2 wt % increases the operation time from 75 seconds at 66°C to 200s at 63°C as well as increasing the thermal conductivity by 1.82 times for the AS5 thermal paste with 2 wt % CNT. Furthermore, CNT TIM pastes with less thickness have a lower thermal resistance of 0.4 K/W. However not all these parameters have been tested with computer chips. This means that in order to increase current heat transfer efficiency limit, we must integrate these parameters into experimental models.

Keywords: Thermal Interface Material; Thermal Paste; Carbon Nanotubes; Thermal Transfer Efficiency; Integrated Circuit; Heat Sink; Heat Dissipation.

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1. Introduction

Integrated Circuit (IC) processors generate significant heat during operation, which degrades performance and will lead to component damage. IC's need to be cooled to prevent the build-up of heat, and as processors become more powerful and smaller and more densely packed, cooling solutions need to improve to keep up.

Cooling of computer chips involve conducting the heat away from the die into a heatsink, which has a large heat capacity and surface area, and using a fan to force airflow over it to dissipate the heat. The cooling is limited by heat capacity, conductivity and ambient temperature of the air. Liquid cooling functions in the same way, however, water pipes are used as the medium to conduct heat away from the die to the heatsink. The high heat capacity of water allows for faster absorption of heat from the die compared to metal mediums.

There have also been developments of evaporation cooling for liquid-based cooling solutions. Rather than water conducting heat to a heatsink, the water is evaporated and then condensed in order to dissipate heat before cycling back to the IC [1]. This method is more suited towards portable handheld devices, as the water cycle is self-contained and does not include moving parts or loud fans. Samsung has been using "water carbon cooling system" in their devices since the Galaxy S7 [2].

Of the stages of heat flow involved in cooling microprocessors, the heat transfer between the die and heatsink is considered the limiting process. The surfaces of the components are non-conforming and rough, resulting in the actual contact area between the two components being substantially smaller than it would appear. The heat transfer between the die and heatsink occurs through micro-contacts and air in the micro-gaps which significantly increases thermal resistance. To reduce thermal contact resistance of the imperfect surfaces, thermal interface materials (TIMs) are used to fill in the micro gaps on the surface to get complete contact. For example, the thermal conductivity of a 25μm thick CPU (silicon)/heatsink (aluminium) interface is 0.357W/mK. When Thermflow T766 is used in the same setup, the thermal conductivity is 0.56W/mK [3]. As TIMs (e.g. Arctic Silver ~7.5W/mK) [4] have lower thermal conduction than copper (~413 W/mK) [5]. The heat conduction rate away from the die is limited and is the reason why TIMs are only applied as a thin layer. Due to the importance of TIM in the overall cooling efficiency of electronic cooling systems it is the focus of this meta-study.

Another method to improve contact area between computer processors and the heatsink involves microfluidic channels being built into a die to allow water to flow directly through the IC. As dies are getting smaller and are beginning to be stacked, it limits the contact area most cooling solutions have between the IC and heatsink as only the top surface of one die in the stack will be in contact. Microfluidic channels, however, still have very good contact with the die as they are built into each individual die in the stack [6]. Although microfluidic channels have high cooling rate implications, they do not use TIMs and so go beyond the scope of this meta-study.

1.1 Conventional Thermal Grease

Common TIMs have a polymerizable liquid medium with a suspension of thermally conductive, but electrically insulating filler. Matrix materials are made of a polymer such as silicone. Fillers are generally made of ceramics such as zinc oxide, alumina, aluminium oxide, silicon dioxide and beryllium oxide [7]. Thermally conductive metals such as silver (403 W/mK, [5]) can be used as the filler, however they can be electrically conductive which can cause malfunction. Polymer matrix thermal pastes are the most common on the market due to their price, performance and ease of application. They have good surface contact and are unlikely to spill out of the heat spreader/die couple. The performance of polymer grease is limited by the conductivity of the filler, conductivity of the matrix and the concentration of filler in the matrix.

Better performance is available in liquid metal thermal pastes (e.g. Thermal Grizzly Conductonaut has a thermal conductivity of ~73W/mK) [8], which use variations on gallium-based alloys. It is more difficult to apply a liquid metal paste as they have low viscosity and so are prone to spillage, this can lead to a malfunction of the IC from short circuits as it is electrically conductive. Gallium is also corrosive to aluminium, which means liquid metal pastes cannot be used together with aluminium heatsinks.

1.2 Carbon Nanotubes

Carbon Nanotubes are an allotrope of carbon which is part of the fullerene structural family. CNTs have the form of graphene rolled into a cylinder to form a “tube” shape. CNTs can form as individual nanotubes known as a single-wall (SWCNT), or as multiple nested nanotubes known as multi-wall (MWCNT). CNTs have many unique and extraordinary properties including incredible thermal conductivity. This makes CNTs a very valuable material for use in TIMs.

Theoretical simulations on carbon nanotubes have produced results indicating that individual carbon nanotubes (CNTs) have a thermal conductivity in the range of 2000 to 6000 W/m K [9]. Due to the difficulty of direct measurement of individual CNTs, there has been a wide spread of results found by researchers. CNTs can be produced as SWCNTs or MWCNTs and their thermal properties can differ significantly. One issue with CNTs is that they tend to form into bundles, this is not ideal as the conductivity between CNT walls is in the range between 67 and 137 W/mK [9]. Due to high thermal contact resistance between tubes, mats of tangled nanotube bundles were found to have much lower conductivity, ~35W/mK for SWCNT and ~15-25W/mK for MWCNT [9]. This means that alignment has a major impact on the thermal properties of CNTs. Another issue with CNTs in a polymer matrix is the interfacial thermal resistance which is caused by phonon mismatch. Gaps between adjacent tubes will cause severe phonon scattering which hinders phonon transport.

Even at the low end CNTs have much greater conductivity than current TIMs and have incredible potential. However, it is difficult to produce CNTs, especially vertically aligned, and their thermal performance is hindered by the interface with matrix materials.

A vertically aligned CNT film can be formed on SiC by a surface decomposition method to obtain properties such as high-density, well-aligned, catalyst-free, flexible CNT tips, high thermal conductivity and high adhesive strength, which are features that meet the requirements of a TIM [10].

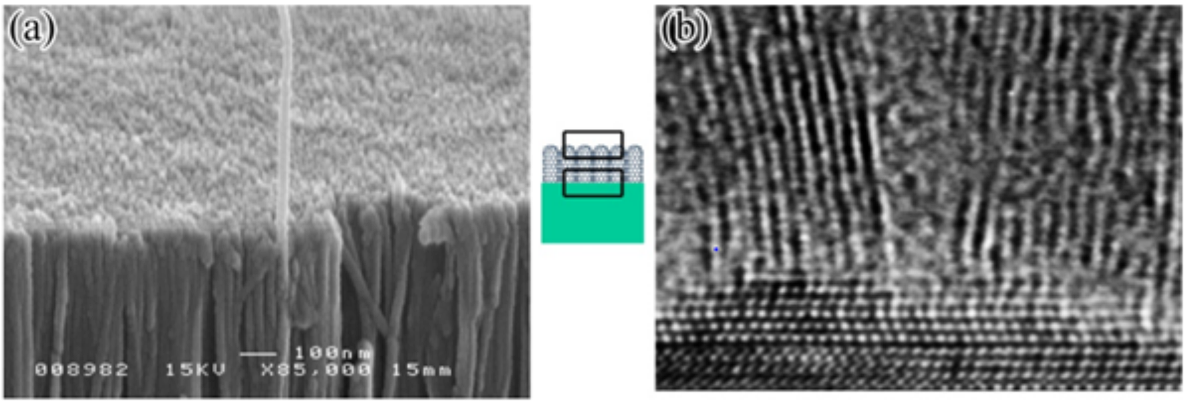


Figure 1. (**a**) Scanning electron microscope image of CNT tips. (**b**) Transmission electron microscope image showing the interface between CNT and SiC [10]

The planar density of CNTs obtained by surface decomposition of SiC is approximated to be $3x10^4\mu m^{-2}$, which is hundred times as high as that of CNTs grown by the CVD method. The high-resolution TEM image shows the presence of (OOO_2) graphite lattice fringes perpendicular to the SiC as shown in Figure 1(b). These fringes are the walls of the multiwalled carbon nanotubes and therefore show that CNTs can easily form onto the SiC substrate at the atomic scale [10]. As well as aligned CNT bundles being closely packed and well aligned have a diameter of approximately 30nm in Figure 1a.

2. Methods

The meta-study draws on literature concerning thermal interface materials and their relation to the cooling of computer chips. Sources were accessed via SCOPUS, IEEE Xplore, Google Scholar and Science Direct. At first the research was directed towards computer cooling in general to achieve a broad understanding of the topic. The object was to then to find relevant data and insight in order to narrow down the broad topic into a single focus point. There was an emphasis on material produced since 2009, ensuring the relevance of the data collected due to rapid advancements in Technology. The keywords which helped us obtain suitable data included ‘thermal pastes vs carbon nanotubes’, ‘thermal interface materials for computer cooling’, and ‘carbon nanotubes and computer cooling’.

The sources used for this meta-study were from the scientific articles and journals mentioned above specifically as independent sources offer an element of reliability and relevance. The articles that were focused included the significant properties that a heat transferring interface material required to achieve its purpose. Articles which focused on comparisons between now-existing polymer matrix thermal pastes and potential carbon nanotube-based thermal pastes were targeted. We investigated scientific journals concerning the thermal properties of carbon nanotubes and how they can be enhanced by mixing different metal alloys and polymer composites to provide maximum conductivity. Because this meta-study doesn't focus on comparisons of existing thermal pastes, we excluded numerous articles based on thermal pastes.

We focused on CNT enhanced TIMs that have improved thermal properties. We collected data on the effect of CNT concentration on temperature increase time of a computer at full load, thermal resistance and thickness of different samples and composites of CNTs and the extra lifetime percentage of LED chips with increased concentration of CNTs in nanofluids. After highlighting the different parameters, we focused on the integration of all them with computer chip and concluded that not all the parameter had been directly tested with computers and therefore, there was a need for further research and experimentation.

3. Results and Discussion

3.1 CNT/SiC composite

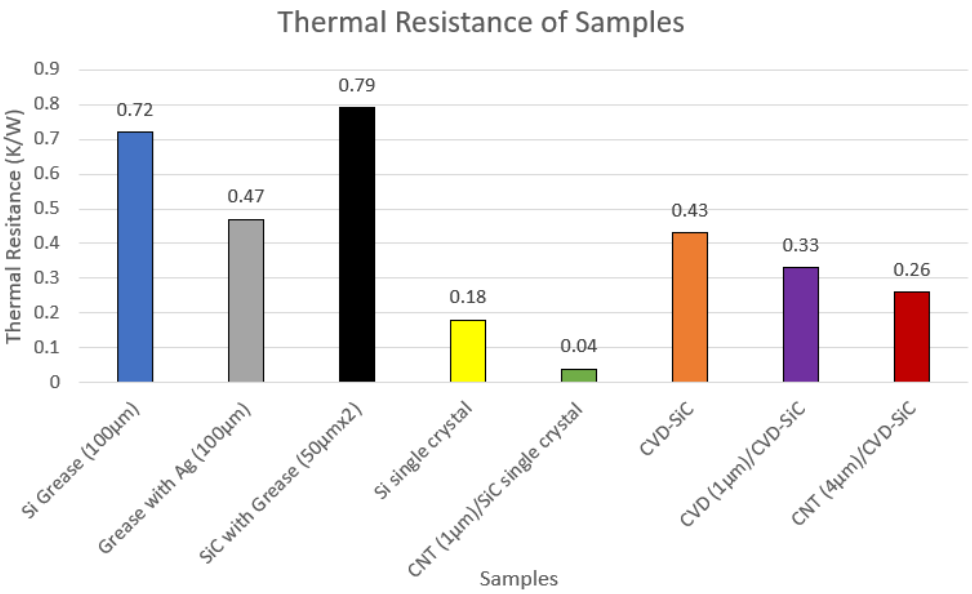


Figure 2. Thermal resistances of various samples tested [10].

In Figure 2, the thermal resistance of different samples created are displayed for an applied pressure of 3750 g/cm^2. The thermal resistances of Si grease and grease with Ag particles were measured to be 0.72 and 0.47 K/W, respectively. The SiC single crystal was found to have a lower thermal resistance of 0.18 K/W, which already shows promise as a potential TIM. This corresponds to a thermal conductivity of 360 W/mK, comparable to copper (403 W/mK) or silver (428 W/mK).

In order to demonstrate the difference thickness can make, a single SiC crystal was coated with silicon grease of thickness 50μm compared to a thickness of 1μm [10]. The thermal resistance measured were 0.79 K/W and 0.04 K/W, suggesting that smaller thickness is largely more effective. The materials with CNTs, without CNTs and single crystal SiC were then compared between CVD method (CVD-SiC), 1µm-CNT/CVD-SiC and 4µm-CNT/CVD-SiC and produced results of 0.43, 0.33, 0.26 K/W respectively [10]. This demonstrates that the resistance of CVD-SiC is higher than that of single crystal SiC because of thermal scattering due to the presence of grain boundaries, however the resistance can be reduced by forming CNTs onto SiC. From these results, it was then concluded forming CNTs on both single-crystal and polycrystalline SiC leads to a high-performance TIM.

3.2 Vertically Aligned Carbon Nanotubes

A vertically aligned carbon nanotube (VACNT) array consists of high-density and well-aligned CNTs which are perpendicular to a substrate. A key parameter for high efficiency of TIMs is a low thermal resistivity which considers the intrinsic thermal resistance and contact thermal resistance between the TIM itself and the contacting material [11]. The total thermal resistance can be divided into three components and is expressed by the equation

$$R_{TIM} = R_{cl} + \frac{BLT}{\lambda_{TIM}} + R_{c2} \tag{1}$$

where R_{C1} and R_{C2} are the thermal contact resistances at the interface between the TIM and the two substrates. λ_{TIM} is the intrinsic thermal conductivity of the TIM and BLT is thickness of the TIM [11]. From the Equation 1, a high intrinsic thermal conductivity of the TIM and a low thermal contact resistance is essential for a high performing TIM. It was observed that the intrinsic thermal conductivity of a VACNT array was determined by both the thermal conductivity of individual CNTs and their packing density [11].

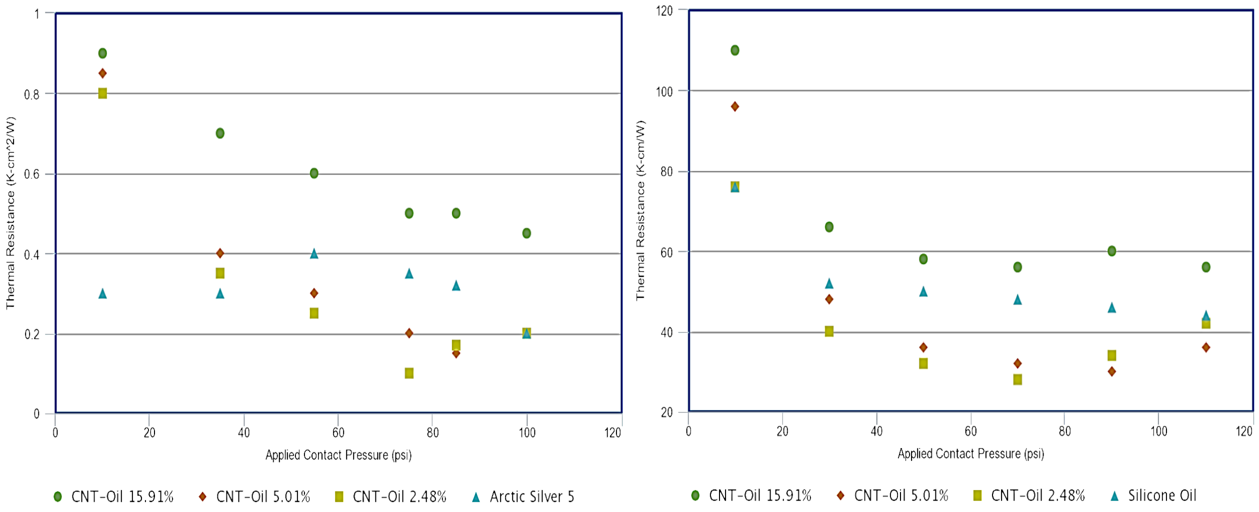
3.3 Optimising Thermal Properties

In order to develop a high-performance TIM, its intrinsic thermal conductivity needs to increase. In order to improve this particular parameter, a suitable growth temperature (700-800ºC), low pressure, specific carbon precursors such as C_2H_2 and C_2H_4, active catalysts (Fe, Co, Ni) is required and the introduction of weak etchants such as water were key factors in obtaining a high intrinsic thermal conductivity of VACNT arrays [11]. Additionally, a smaller diameter results in a higher thermal conductivity. The diameter is controlled by tailoring the catalyst film thickness or inhibiting the aggregation of the catalysts by increasing the interaction with a buffer layer or coating it with a thin inert layer [11].

A method of increasing thermal conductivity and decreasing thermal contact resistances at interfaces of VACNTs is by ‘thin carbon covering’ which smooths out the rough surface of the VACNT arrays (Figure 6). This technique also connects the free tips of the VACNT array and therefore ensures that most nanotubes in the array touch the interface surface and allow for heat transport. Thermal resistance of CNTs decreased from 40mm^2 KW^{-1} to a very low 1.3mm^2 KW^{-1} with a coating thickness of 20µm [11]. This process is very simple, and a wide working temperature range is achieved due to excellent thermal stability of the carbon covering, hence decreasing the overall thermal contact resistance.

3.4 Carbon Nanotubes with Silicone Oil

A set of interface materials with CNTs mixed into silicone oil, includes a high percent weight concentration of CNTs [12]. The CNT-oil mixtures show better total thermal resistance when compared to CNTs without oil and an increase in pressure.



(**a**) (**b**)

Figure 4. (**a**) Total thermal resistance per unit surface area of Arctic Silver 5 and CNT TIM mixtures over applied pressure range 7. (**b**) Total thermal resistance per unit surface area of Silicone Oil and CNT TIM mixtures over applied pressure range [12].

In Figure 4a, the CNT-oil mixtures show better total thermal resistance than the Arctic Silver thermal paste. Similarly, it also shows significant reduction in overall resistance as pressure increases. For example, two of the CNT/Oil mixtures at lower CNT fractions (2.48% and 5.01%) demonstrates an overall improvement on the thermal paste at higher pressures.

In Figure 4b, the total resistance is divided by the sample thickness and shows a strong collapse when approaching a constant value. At lower pressures, the resistance is greater indicating that there is an increase in conductivity of the mixture, which leads to an increase of surface wetting of the interface material, hence lowering the value of resistance at high pressures. The CNT-Oil mixtures display a better collapse among the different CNTs, implying the thermal resistance depends more on the interface thickness and less on the thermal conductivity. The resistance test results indicate a variation of a factor of 7, while the resistance over thickness measurements differ by 50% [12]. When comparing the CNT-oil mixture to the commercial Arctic Silver thermal paste, it is discovered that the thermal conductivity and the improvement in interface resistance is due to a reduced thickness of the CNT. The values of resistance for these interface materials show that the inclusion of CNTs shows an improved thermal conductivity. As a result, a mixture composed of CNTs and silicone oil yields an improved performance on the commercial thermal paste. The CNT/Oil also shows high compliance and reduction of total resistance with the increase of pressure.

3.5 CNT in microprocessors and LEDs

The addition of CNTs to thermal pastes in microprocessors have shown to increase the operation time, implying the time it takes for the temperature of the microprocessor to reach a constant temperature. Nguyen, Bui and Phan observed the effects of two different thermal pastes containing CNTs (a silicon thermal paste Stars and an AS5 thermal grease which used multi-walled CNTs) on the operation time and temperature of a personal computer (Intel Pentium IV) working at full load [13]. Figure 5 shows the measured temperature of the microprocessor as a function of operation time over concentrations of CNTs ranging from 0-7% for the Stars thermal paste [13]. When no thermal paste is used, the computer heats up to 85°C and shuts down in 20 seconds [13]. As the CNT concentration was increased in the Stars thermal paste, the temperature of the microprocessor decreased gradually. It can be observed that with 2wt% CNT, the temperature increase time and saturated temperature of the microprocessor was 200s and 63°C respectively (most efficient) [13]. In contrast, the Stars thermal paste without CNT had values of 75s and 66°C respectively [14]. The same pattern was observed for the AS5 thermal paste.

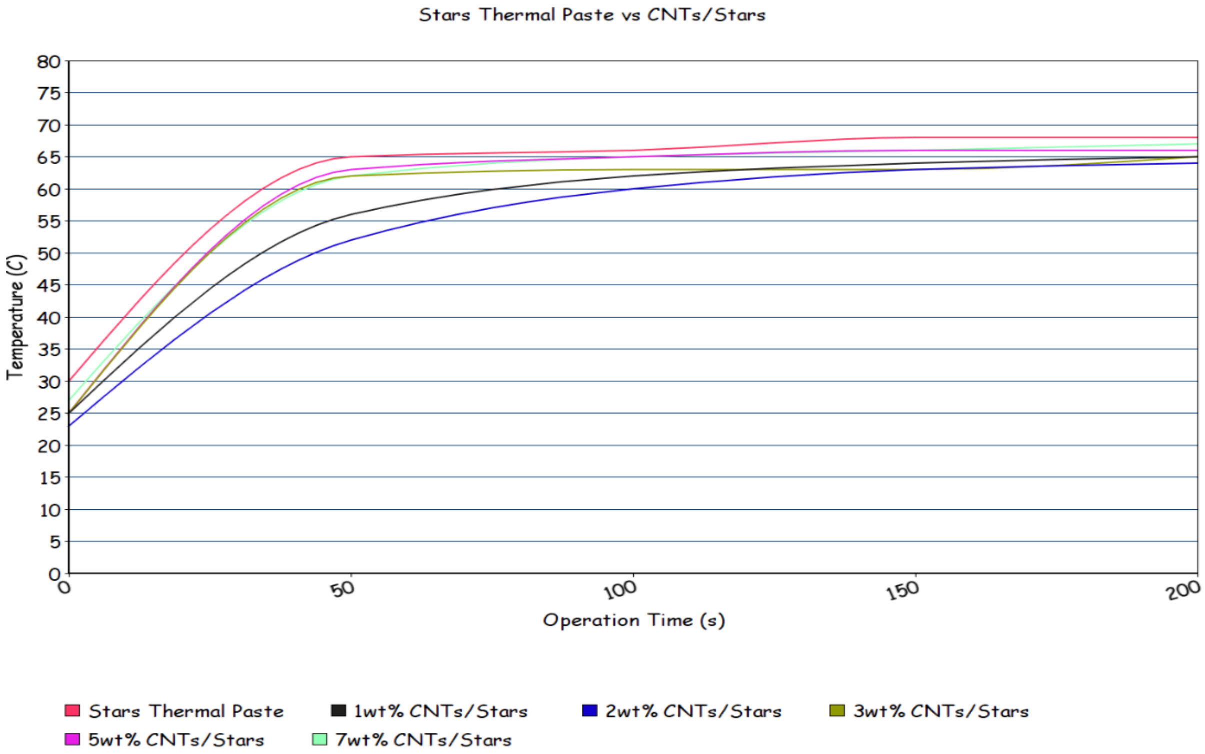


Figure 5. Temperature of the micro-processor as a function of working time using different thermal greases: Stars, 1 wt% CNTs/Stars, 2 wt% CNTs/Stars, 3 wt% CNTs/Stars, 5 wt% CNTs/Stars and 7 wt% CNTs/Stars thermal grease [13, 14]

Table 1 shows the effect CNT have on thermal conductivity and thermal resistance. The results clearly confirm that an increase in CNT concentration increases the thermal conductivity and decreases the heat resistance of the thermal paste. For the case of Stars thermal paste with 2% CNT, the thermal conductivity was 1.37 times higher than the thermal paste without any CNT. Similarly, the AS5 thermal paste with CNT had a thermal conductivity of 1.82 times higher than the thermal paste without CNT [13]. The thermal conductivity was calculated using the equation

$$K_{CNT/Stars} = \frac{t}{R_{th} \times A} \tag{2}$$

where K is the thermal conductivity, t is the thickness of the thermal paste, A is the area covered by the thermal paste and R_{th} is the thermal resistance. [13, 14, 15]

Table 1. Thermal conductivity and thermal resistance of Stars and AS5 at 0 and 2 wt % CNT

CNT concentration	**Thermal conductivity W m^{-1} K^{-1}**	**Thermal Resistance K W^{-1}**
Stars Thermal Paste 0wt% CNT	1.87	0.13
Stars Thermal Paste 2wt% CNT	2.56	0.095
AS5 Thermal Paste 0wt% CNT	8.89	0.027
AS5 Thermal Paste 2wt% CNT	16.2	0.015

Table 2. The effect of increase in concentration on temperature and extra life time percentage on Nano fluid in LED floodlight chip

CNT Nano fluid concentration gram/l	**Temperature °C of LED chip**	**Extra life time %**
0.3	55.0	10.0
0.5	53.7	18.2
0.7	52.5	24.9
1.0	51.9	32.0
1.2	50.6	32.5

Table 2 shows the extent of the effect CNTs have on the lifetime of an electronic device, in this case a 450W LED flood light. The table clearly shows as the concentration of the CNTs in the nano fluid of the LED increased, the temperature of the LED chip decreased and further extended its lifetime. The extra lifetime percentage was determined by the expression

$$L\% = \frac{L - L_o}{L_o} \times 100\% \tag{3}$$

where L_0 is the basic lifetime and L is the extra lifetime [13]. Although this doesn't apply directly to computer chips, the results are promising and can further be tested under the same parameters on computer chips.

4. Conclusions

This meta-study has assessed several enhancements that can be made to CNT TIMs to improve their efficiency. Specifically, these enhancements include a decrease in thickness of CNT/SiC composite, increase in the intrinsic thermal conductivity, decrease in the thermal contact resistance of VACNTs and an increase in the concentration of CNTs used. The primary focus of this meta-study was on the suitability of using CNTs in TIMs for use with computer chips. The presence of CNTs in thermal pastes proves to greatly increase the time it takes for the temperature of a microprocessor to reach a constant temperature. with CNT concentration of 2 wt % in a thermal paste (Stars and AS5) it takes a microprocessor 200s to reach 63°C.

The results clearly outline the boundaries at which maximum thermal efficiency can be reached. Single SiC crystals with 1μm thickness of thermal grease proved to harbour the lowest thermal resistance, 0.04 K/W. In addition, decreasing the thermal contact resistance by thin carbon covering proved to decrease the thermal resistance profoundly from 40mm^2 KW^{-1} to 1.3mm^2 KW^{-1}. We can therefore conclude that not all the parameters listed above have been tested on computer chips. The concentration of CNTs was the only parameter we found that had been tested and we believe that the other parameters (thickness, thermal resistance, intrinsic thermal conductivity) we investigated need to be integrated into further experimentation models with computer chips. If all these parameters are tested individually with computer chips, and later hybridized into a single TIM, an exceptionally efficient TIM could be manufactured. This therefore warrants the need for further investigation and experimentation.

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