Graphene and Carbon Nanotubes for Electronics Nanopackaging

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In recent years, the aggressive downscaling of electronic components has led to highly dense and power-hungry devices. With Moore's law expected to soon reach its physical limit, there is a pressing need to significantly improve the efficiency and performance not only of nanodevices, but also of the embedding environment in which such nanodevices are integrated. In this context, key for improving the performance and for reducing both system cost and size is electronics packaging. However, electronics packaging at the nanoscale (i.e., nanopackaging) is currently facing several technological challenges, as in such scale conventional materials present intrinsic physical limitations. To address this, it becomes necessary to replace these latter with novel alternatives, such as low-dimensional carbon-based nanomaterials. Carbon nanotubes (CNTs) and graphene (materials with 1D and 2D dimensionality, respectively) have the potential to be successfully integrated into traditional silicon-based electronics as well as with beyond-silicon electronics, and their unique electrical, thermal, mechanical, and optical properties could be key enablers for significant performance improvements. In this short review we describe why these nanomaterials are very promising for electronics nanopackaging, and we outline the key application areas, mainly interconnects, thermal management, and flexible devices.

Index Terms—Carbon Nanotube, Flexible Electronics, Graphene, Interconnects, Nanopackaging, Thermal Management

I. INTRODUCTION

THE rapid development of nanoelectronics technology has enabled the high integration of transistors and the significant miniaturization of electronic devices. From a technological point of view, such outstanding progress has allowed integrated circuits to continuously satisfy Moore's law, at the cost of facing unforseen technological difficulties. In addition to the inherent physical limitations of device downscaling, assembly and packaging are becoming the bottleneck factors in the performance of systems, at different levels [1] (see Fig. 1).

Nanopackaging plays a crucial role for the future of nanoelectronics, as outlined in the IEEE EPS Heterogeneous Integration Roadmap (HIR) [2]. Among the key technological areas discussed therein, flexible electronics, interconnects, and thermal management have attracted increasing interest, as can be seen by the sharp increase in the number of scientific publications on such topics over the past 20 years (see Fig. 2).

Perhaps one of the greatest challenges in nanoscale devices is the thermal management: the rising density of transistors per volume area has caused a dramatic increase of the heat flux and the power dissipation in devices, and therefore packaging materials are required to possess (among others) good thermal properties for the efficient removal of heat. Another significant challenge is posed by the physics of materials in the "nano" domain, whose properties may greatly vary with respect to the "micro world". For instance, conventional interconnects made of copper are known to suffer from a sharp increase in the resistivity as their line width approaches few tens of nanometers (typpically < 40 nm). This effect is due to both grain boundary and surface scattering. Another issue
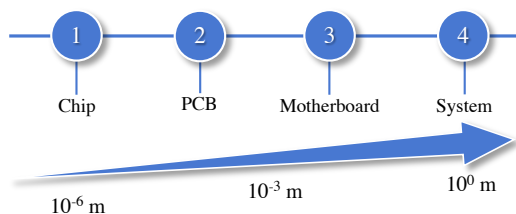

Fig. 1. Hierarchy of the four fundamental levels of electronics packaging.
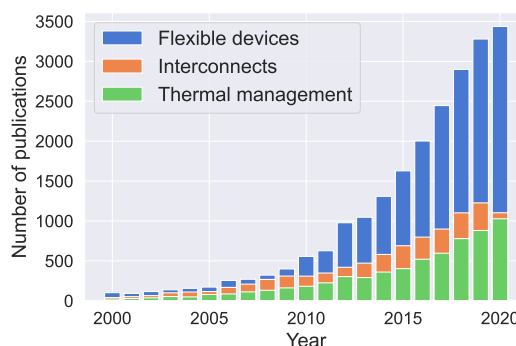

Fig. 2. Number of scientific publications related to flexible electronics, interconnects, and thermal management with carbon-based materials, between the years 2000 and 2020 (source: Web of Science).

is the increased electromigration caused by the higher self-heating. Finally, the rise of flexible and wearable electronics technology, especially for medical and healthcare applications, requires novel heterogeneous system-in-package integration strategies. If the aim is to obtain non-invasive wearable devices, packaging and support materials (e.g., interconnects, wiring and components) need to take into account conformability, flexibility, and biocompatibility.

It is clear that conventional packaging materials may need to be replaced with novel and more advanced alternatives. To this end, research has been heavily focusing on carbon-based materials such as carbon nanotubes (CNTs) and graphene, as they both possess desirable electrical, thermal, mechanical, and optical properties. Thus, the integration of this class of mate-
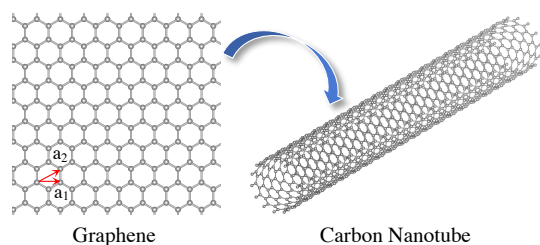
Fig. 3. Chemical structure of graphene and single-wall carbon nanotubes. The roll-up vectors $\mathbf{a}_1$ and $\mathbf{a}_2$ are also shown in red.

rials into devices has the potential to enable new architectures, significant performance improvements, and novel applications.

The present review aims at providing an overview on carbon materials (mainly CNTs and graphene) and their properties, and will discuss their recent applications in the context of electronics packaging. This paper is then organized as follows: Section II (Materials and Properties) will provide a brief description of the electrical, thermal, mechanical, and optical properties of such materials, highlighting what makes these intriguing to be used for nanopackaging. Section III (Applications of CNTs and Graphene in Nanopackaging) will focus on some of the most technologically relevant applications of carbon materials in nanopackaging, mainly interconnects, thermal management, and flexible devices. Finally, Conclusions are discussed in Section IV.

II. MATERIALS AND PROPERTIES

This section is dedicated to the description of the relevant properties of carbon materials (i.e., graphene and CNTs) for their use in electronics packaging applications. Here, we will mainly focus on their electrical, thermal, mechanical, and optical characteristics, and we will elucidate how these are determined by their chemical structures, both shown in Fig. 3.

Graphene and CNTs are low-dimensional materials and both allotropes of carbon. The former is the 2D counterpart of bulk graphite, and consists of a flat sheet of C atoms in a hexagonal (or honeycomb) arrangement. In such a lattice, each C atom forms strong σ bonds with the three adjacent C atoms via sp^2 hybridization of the carbon orbitals, whereas π-conjugation occurs in the out-of-plane direction. Graphene can be seen as the building block of CNTs, whose C atoms essentially retain the same chemical connectivity of their "precursor" material. CNTs are rolled-up sheets of graphene of cylindrical shape (hence, the 1D dimensionality), and the curved π-conjugation of the C atoms in the lattice determines their unique chemical/physical behavior [3]. Conventionally, CNTs are classified based on their chirality, which is uniquely defined by the roll-up vector:

$$\mathbf{C}_h = n\mathbf{a}_1 + m\mathbf{a}_2. \qquad (1)$$

Here, $\mathbf{a}_1$ and $\mathbf{a}_2$ are the primitive vectors of the graphene sheet (see Fig. 3), whereas n and m are integers. Notably, the electronic properties of such class of materials largely depend on their chirality as well, as will be later outlined. Moreover, CNTs can be either single-wall (SWNTs) or multi-wall (MWNTs), if made of concentric cylindrical shells.

Graphene, with its characteristic electronic band structure, is a zero-gap material. On the other hand, nanotubes can show either semiconducting or metallic behavior, depending on the integer values n and m: in general, if $n = m$ the CNT is metallic, otherwise it is semiconducting (or, in some cases, semi-metallic). This can be very well explained theoretically by the band theory and the zone-folding approximation [4].

Since the discovery of both graphene and CNTs, several theoretical and experimental studies have been carried out to understand, predict, and determine their unique properties. However, before delving deeper into what makes both materials very attractive for packaging applications, it is important to provide a brief description of their synthethic routes and growth mechanisms, as ultimately the materials' properties are deeply affected by their structural quality.

A. Synthesis of Graphene and CNTs

Graphene was first obtained in 2004 by mechanical exfoliation of highly oriented pyrolitic graphite, and the process is described in detail in the well-known work by Novoselov et al. [5]. Unfortunately, mechanical exfoliation only allows to obtain small (but high quality) graphene flakes of the size of few tens of μm, with irregular shapes and with no control on their orientation. This is clearly a disadvantage: on the one hand, if the aim is mass production, it becomes necessary to obtain large-scale graphene sheets; on the other hand, if the aim is to fully exploit the material's electronic properties, better control on the flakes orientation and on the material quality is needed.

Nowadays, graphene can be obtained via several methods, either by direct growth from a precursor (bottom-up approach) or by exfoliation of graphite (top-down approach). Among the bottom-up methods, chemical vapor deposition (CVD) and epitaxial growth on silicon carbide (SiC) are widely used, although recently alternative procedures to obtain high yields of graphene have been explored [6]. CVD is particularly well suited for mass production, and it involves the formation of graphene on a metal substrate by exposing such substrate to a precursor gas, often methane, at high temperatures ($\sim 1000°C$). The metal substrate is mostly Cu, although Ni and Pt have also been used. The drawback of this process is the fact that graphene, once obtained, needs to be transferred onto an insulating substrate, and this causes the formation of structural defects, tearing, and the crumpling of the sheets. Epitaxial growth on SiC is a promising growing method for graphene. Essentially, this process involves annealing of hexagonal SiC crystals at high temperatures ($> 1000°C$) and in ultra high vacuum. The crystal structure of SiC determines the quality of the graphene layers, and the sheets produced via this method are generally uniform and structurally coherent. The main drawback of this process is the high cost, which is derived from the extreme growth conditions of the material.

Among the top-down approaches, liquid-phase exfoliation (LPE) [7] of graphite and/or graphite oxide is the most used synthetic route to obtain graphene flakes. This method involves the immersion of graphite in organic solvents, such as N,N-dimethylformamide (DMF) and N-methyl-pyrrolidone

TABLE I
COMPARISON OF THE UPPER LIMIT OF THE ELECTRICAL, THERMAL, AND
MECHANICAL PROPERTIES OF CNTs AND GRAPHENE AGAINST
STATE-OF-THE-ART COPPER CONDUCTIVE MATERIAL.

	Graphene	CNTs	Cu
Elec. conductivity (S cm^{-1})	$\sim 10^5$	$\sim 10^6$	$\sim 10^5$
Ther. conductivity (W m^{-1}K^{-1})	5300	> 6000	385
Max. current dens. (A cm^{-2})	10^8	10^9	$< 10^7$
Young's modulus (TPa)	1	1	0.13

(NMP), in order to weaken the interlayer van der Waals forces by solvent intercalation. This process is then followed by ultrasonication. Alternatively, graphite can undergo the initial oxidation step to produce graphite oxide, which will then get exfoliated in solution to produce flakes. The flakes will then need to be reduced to recover graphene.

Similarly to graphene, different techniques have been used to produce CNTs, mainly electric arc discharge (which led to the first discovery of this class of materials [8]), laser ablation, and CVD. The first two methods are generally rather expensive, and therefore not suitable for mass production, although CNTs produced via these processes present generally very little structural defects. What makes both arc discharge and laser ablation unsuitable for mass production are the extremely high temperatures, $> 1700°C$ and $> 1200°C$, respectively. On the other hand, CVD requires lower temperatures ($\sim 700°C$) but a longer reaction time, and this leads to more structurally defective CNTs. As for graphene, CVD requires the growth of CNTs over a metal substrate, generally Ni, Co, Fe, and the precursor gas is once again often methane.

B. Electrical Properties

The unique transport properties of graphene and CNTs derive from their chemical structure (sp^2 bonds, as described earlier), and the quantum confinement given by their intrinsic 1D and 2D geometries.

In the case of ideal free-standing graphene, its carrier mobility can reach extremely high values, up to 2×10^5 cm^2 V^{-1} s^{-1} [9]. Moreover, this value shows very little dependence on temperature. However, structural defects and the support material (e.g., SiO$_2$) are known to severely limit the room temperature mobility to $\sim 4 \times 10^4$ cm^2 V^{-1} s^{-1} [10]. In addition, reported resistivity measurements showed very low values, of the order of 0.01×10^{-6} Ω m [10]. The carrier mobility of CNTs have also been estimated to be exceptional and close to 10^5 cm^2 V^{-1} s^{-1} [11].

In the ideal situation, the electrical transport of CNTs can be assumed to occur in the ballistic regime. Assuming perfect ohmic contacts reservoirs, the theoretical upper limit of the electrical conductance (G) in CNTs is proportional to the conductance quantum $G_0 = 2e^2/h$, multiplied by the number of effective quantum channels. For instance, small-diameter metallic CNTs generally present two quantum channels, and therefore $G = 2G_0$ at charge-neutrality point. However, in reality contact resistance at the electrodes significantly lowers G, and therefore good metal contacts need to be explored. Recently, the electrical conductivity of MWNTs was found to be enhanced when in contact with nanoscale Cu electrodes,

but other metals like W and Cr were also tested [12]. On average, several electrical conductivities have been reported for CNTs, depending on the quality of the material, as well as the diameter and the growth process. More specifically, for SWNTs conductivities were reported in the range of 10^2-10^6 S cm^{-1}, whereas for MWNTs in the range of 10^3-10^5 S cm^{-1}. CNT bundles and fibers were found to have yet very different electrical conductivities, ranging between 10 and 67,000 S cm^{-1}. For instance, Park et al. [13] grew CNT fibers by means of CVD, and found a conductivity of 1480 S cm^{-1}, whereas Hossain et al. [14] found a value of 975 S cm^{-1}.

In addition to very good electrical conductivities, CNTs can sustain high current densities ($\sim 10^9$ A cm^{-2}), and this is one of the reasons that makes this class of materials very attractive for interconnects. It is worth noting that also graphene shows very high current density, of the order of 10^8 A cm^{-2} [15]. As a comparison, the standard Cu interconnects can sustain only $\sim 10^6$ A cm^{-2} due to electromigration, thus limiting their applicability in nanoscale devices. Therefore, CNTs are seen as an extremely valid alternative for energy-efficient integrated circuits [16], [17], as will be discussed in the next sections.

C. Thermal Properties

Thermal transport in both graphene and CNTs is largely dominated by phonons, even when the nanotubes are metallic. The specific heat (C) of a material in general depends on both phonon (C_{ph}) and electron (C_e) contributions. Indeed, it has been reported that for the in-plane modes of graphene the ratio $C_{ph}/C_e \approx 10^4$, whereas for SWNTs the ratio is $\approx 10^2$. Similarly to the electrical conductance (G), thermal conductance (G_{th}) of CNTs can be assumed to be ballistic [18], and proportional to the thermal conductance quantum:

$$G_0^{th} = \frac{\pi^2 k_B T}{3h}, \tag{2}$$

where k_B is the Boltzmann constant, T is the temperature, and h is the Planck's constant. In the limit of defect-free and ideal materials, both graphene and CNTs were found to have comparable thermal conductivities: up to 5300 W m^{-1}K^{-1} for free-standing graphene [19], and up to 6600 W m^{-1}K^{-1} for a (10, 10) CNT [20]. Such very large estimated thermal conductivities make both materials particularly attractive for thermal management applications. For instance, this is once again relevant for interconnect applications, as CNTs can significantly outperform Cu in heat diffusion, thus potentially reducing the temperature in the chip.

More recently, graphene- and CNT-based nanocomposites have been extensively explored as thermal interface materials (TIMs). In the context of flexible electronics, nanocomposites are extremely promising in this sense, as they combine the flexibility of the polymer matrix with the high thermal conductivities of graphene and CNTs. For instance, Cui et al. [21] used silica-coated MWNTs within an epoxy matrix, and they were able to demonstrate excellent flexibility, and high electrical and thermal conductivities in the nanocomposite. Shahil et al. [22] developed a graphene-multilayer graphene nanocomposite, and they measured an enhancement of the thermal conductivity of the polymer composite by 2300%.

D. Mechanical Properties

CNTs and graphene are extremely robust to mechanical deformation. In the ideal situation, that is with both materials in their pristine state, they can be considered among the strongest and stiffest materials currently available, in terms of tensile strength and Young's modulus. Their outstanding mechanical properties directly depend on the strength of the sp^2 C-C bonds. A rough estimate of the bond strength can be obtained by considering a simple harmonic model approximation: the Young's modulus related to the C-C bond would be then proportional to the spring constant k. Such value is between 500 and 1000 N/m, whereas for metals and ionic solids it is only $\sim$100 N/m [23].

Theory predicts both materials to have exceptional Young's moduli in the order of $\sim$1 TPa [24], thus comparable to diamond. Early experiments on nearly defect-free CNTs corroborated this: for instance, Krishnan et al. [25] reported an average Young's modulus of $\sim$1.3 TPa, and Yu et al. [26] a mean value of $\sim$1 TPa. However, as previously mentioned, nowadays the growth method of choice is CVD, which allows to obtain very high yields at the expense of the materials' quality. Moreover, CNTs are mostly found in bundles/networks, and their mechanical properties are far from the theoretical ideal case. Also MWNTs show lower Young's moduli with respect to a single pristine SWNT: for instance, Yu et al. [27] reported 18-68 GPa, whereas Kim et al. obtained 34.65 GPa. More recently, Andalouci et al. [28] measured the Young's modulus of vertically aligned MWNTs and they found a value of 850 GPa. Tensile strengths of SW and MW nanotubes were estimated to be $\sim$175 GPa and $\sim$63 GPa, respectively. Clearly, these values are only indicative, as the elasticity of either SW or MW nanotubes and bundles depends on numerous factors, such as the nanotube diameter, chirality, geometric arrangement, intrinsic defects, and growing technique.

Similarly to CNTs, also the mechanical properties of graphene are found to be affected by the quality of the sheet. Lee et al. [29] measured an average Young's modulus of $\sim$ 1 TPa for a free-standing graphene monolayer, however crumpling and point defects can significantly vary this ideal value [30]. On average, the tensile strength of graphene has been measured as $\sim$130 GPa.

It is important to point out that although being far from the ideal values, the mechanical properties of CNTs and graphene are still exceptional. Indeed, both materials are used as reinforcements in nanocomposites, with important applications in wearable health-monitoring sensors [31], [32].

E. Optical Properties

The peculiar electronic properties of graphene, determined by its band structure and its density of states (DOS) in proximity of the Dirac points, also lead to the material's peculiar optical properties. For instance, the absorption of graphene was found to be non negligible and defined only by its fine structure constant (α), which can be expressed as:

$$\alpha = \frac{e^2}{4\pi\epsilon_0 \hbar c}. \qquad (3)$$

Here, e is the elementary charge, ϵ_0 is the vacuum permittivity, $\hbar$ denotes the reduced Planck's constant, and c is the speed of light. What is striking, is the fact that although having one-atom thickness, graphene can absorb $\sim$2.3% of the incident light [33], regardless of its wavelength. Moreover, it was observed that such absorption is linearly proportional to the number of layers.

CNTs also present peculiar optical properties. For instance, in semiconducting CNTs the optical band gap (and consequently the adsorption onset) was found to be inversely proportional to the diameter of the tubes. As a direct consequence, the overall absorption spectrum of a CNT sample could be in principle tuned by selecting a combination of CNTs with specific diameters. The presence of several sharp peaks is typical of the absorption spectra of CNTs, each peak corresponding to one of the electronic transitions (e.g., E_{11}, E_{22}, . . .) [34]. Additionally, semiconducting CNTs are also fluorescent, as these are able to emit light in the near-IR region upon photoexcitation.

Interestingly, both graphene and CNTs were found to have extremely fast saturable absorption recovery times, on the order of picoseconds, which could enable the use of both materials in the field of ultrafast photonics, for instance as fibre lasers [35].

III. APPLICATIONS OF CNTS AND GRAPHENE IN NANOPACKAGING

In this section, we will focus on technologically relevant applications of both graphene and CNTs in electronics packaging, more specifically, interconnects, thermal management, and flexible devices.

The improvement in performance (and cost reduction) of the information and communications technology (ICT) in recent years has been primarily correlated to the increase in packing density of devices over the chip, assisted by the aggressive downscaling of their size. Nevertheless, the efficiency of electronic gadgets is ruled not only by that of micro- and nanodevices, since electronic appliances mostly consist of interconnected assemblies made of individual components. The integrating backbone takes the name of electronics package, and it provides four major functions: electrical interconnection, power distribution, heat dissipation, and mechanical protection. Thus, the performance of the embedding environment deeply impacts in turn the performance of the technological objects. Particularly now that Moore's law is close to reach its intrinsic physical end, a packaging version of it has been invoked [37] as liable to drive, by system scaling, the efficiency improvement of ICT in the next years.

A. Interconnects

On-chip interconnects are organized into a hierarchy: global interconnects, which deliver signal and power, intermediate interconnects, for joining cells to cores/modules, and local interconnects, for cell-to-cell bridging. The global interconnects are the most affected by device scaling, as well as electromigration issues, due to the large amount of current that they have to support. A steep increase in both resistance and
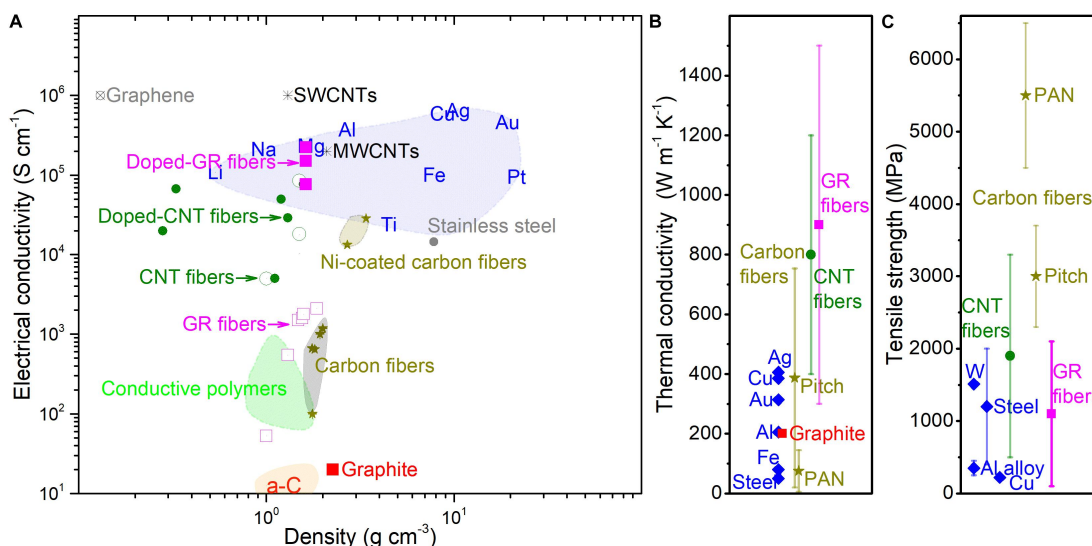
Fig. 4. (A) Electrical, (B) thermal conductivities, and (C) tensile strength of CNT- (dark green) and graphene-fibers (magenta) compared to carbon fibers (brown) and some metals (blue). Original work from [36], reproduced under CC-BY-4.0.

capacitance is the main problem experienced by the scaling of intermediate interconnects. Capacitance effects may be dealt with air gaps, though the increment of resistance remains critical. Finally, local interconnects are also interested by the miniaturization trend, given that the shrinkage of device dimensions is followed by the parallel decrease of wiring size, with detrimental effects on resistance. Thus, on the one hand, the intrinsic gate delay is diminishing, whereas on the other hand, the RC delay due to interconnects becomes dominant. State-of-the-art interconnects are currently implemented in copper, however the accelerated technological scaling is showing their bottleneck factors, represented by 1) the increment of resistivity due to enhanced grain boundary scattering and surface scattering with the shrinking of the Cu width [38]; and 2) the electromigration effect. Therefore, research on alternative conductor materials for the implementation of on-chip interconnects is essential for boosting the performance and the energy efficiency of future chips [16], [17], [39].

As mentioned in the previous section, CNTs possess very desirable properties as the next very large scale integration (VLSI) interconnects, such as high electrical conductivity (Table I). Because one single CNT can carry higher current densities compared to one Cu interconnect (Table I), a few CNTs can carry the same amount of current as a typical Cu interconnect (cross-section 100 nm × 50 nm). Additionally, SWCNTs have shown to possess a coefficient of thermal expansion (CTE) of about $2 \times 10^{-6}/°C$ [40], [41], quite close to that of silicon ($2.3 \times 10^{-6}/°C$). This makes the use of SWCNTs a clear advantage with respect to Cu for the realization of global interconnects for 3D stacking by flip chip bump and through-silicon via (TSV) [41]. Since Cu has a much higher CTE ($17 \times 10^{-6}/°C$) than silicon, then pillar bumps and TSVs based on Cu create stability and reliability problems during chip operation, when integrated into silicon. Finally, CNTs are very efficient heat spreaders due to their much larger thermal conductivity compared to Cu (Table I). However,

many challenges remain to be faced for CNTs to become the novel on-chip back-end-of-line (BEOL) interconnects. In this respect, simulations of CNTs can be a valuable tool for the research and development step, complementary to experiments.

Simulations based on first-principle methods have been extensively carried out to model electrical and thermal properties of CNTs [42], [43], [44], [45]. The development of compact models of CNT interconnects, suitable for circuit-level simulations, is of particular importance for providing guidelines for a future technology based on CNT interconnects. The development of a compact model of SWNT-bundle interconnects [46] has allowed to assess their performance by taking into account realistic issues, such as their contact resistances or the lack of control on chirality, as well as the (local, intermediate or global) domain of interconnections. Compared to SWNTs, MWNTs present advantages, such as the fact that their growth can be better controlled. Also, MWNTs are always metallic, whereas the kind of semiconducting or electrical conductivity of SWNTs depends on their chirality. An equivalent distributed circuit model for MWNT interconnects [47] has enabled their comparison with Cu and SWNT interconnects. It has been found that there is a sizeable improvement of signal delay for global- and intermediate-level MWNT interconnects, but not for short local interconnects, if compared to Cu vias. Additionally, MWNT interconnects can perform better than SWNT bundles aflicted by random chiralities or low densities. A hierarchic approach, from atomistic- to circuit-level, has been used to investigate the trade-offs on SWNT interconnect performance between defects, delay/contact resistance and doping [48]. A compact shell-level model of MWNT has been developed to take into account defects, chirality, end-contact resistance, and shell connectivity to end contacts [49]. This model has been then applied to study the impact of charge transfer doping [50]. Moreover, it has been used to explain the efficiency of Pt-salt-doping as a strategy to reduce MWNTs

resistivity by converting semiconducting shells to metallic ones, as well as its potential as a solution to balance the effect of defects and contact resistances [51], [52].

The family of "all-carbon" electrical conductors comprises also fibers, yarns and ropes obtained by assembling CNTs and/or graphene. A recent and extensive review can be found here [36]. CNT/polymer and graphene/polymer fibers are applied in fields such as electromagnetic shielding, wearable and smart textiles or as electrodes. However, they do not achieve the thermal and electrical conductivity of metals, on the contrary of nanocarbons assembled into fibers, whose quality is however highly dependent on the fabrication process. CNT yarns can be obtained by dry-spinning procedure or twisting during spinning. CNT and graphene fibers can be also achieved by electrospinning or wet spinning procedures. The conductivity of CNT fiber conductors depends from multiple factors, such as doping, purity and density of the CNTs [53], where the electrical resistivity in the parallel direction can be orders of magnitude smaller than that along the orthogonal direction. Assemblies of CNTs usually possess resistivities two order of magnitude larger than single metallic CNTs, and disordered mats of CNTs have demonstrated twenty times larger resistivity [54]. This is indicative that the contact resistance at the contacting interfaces between single CNTs has a major role [55]. In particular, theoretical investigations have demonstrated the angular dependence of conductivity between two SWNTs in contact, which in turn varies according to the fact that the SWNTs have the same or different chiralities [56]. An overview of the electrical and thermal conductivities, and tensile strength of CNT and graphene fibers is shown in Fig. 4 [36].

B. Thermal Management

Closely related to the scaling of devices (and associated interconnects) is the issue of thermal management, which in the past was a post-chip-making issue. Nowadays, it is necessary to deal with it starting from the packaging level of device and circuits. The increase in packing density of devices over the chip (while the die size stayed relatively constant), has implied, on the one hand, an equally significant increase in both the power and thermal densities (see Fig. 5). On the other hand, the shrinking of interconnects size has led to an increase of their thermal resistance, with consequent difficulties for the removal of heat [57]. Effective cooling strategies are mandatory not only for reliability and longevity of nanoscaled devices, but also for other aspects, such as the avoidance of overheating of batteries. Progress in mobile communications, smart appliances and automotive industry are all made possible by high-performing batteries, such as Lithium-ion (Li-ion) ones. However, fast charging/discharging processes or short-circuiting may cause cell rupture or even explosion, with consequent health and environmental risks. Thus, materials with high thermal conductivities can boost the transfer rate of heat from devices and circuits, and are ideally suited as heat spreaders. Hence, heat diffuses more efficiently through CNT than Cu vias, and this can reduce the on-chip temperature.
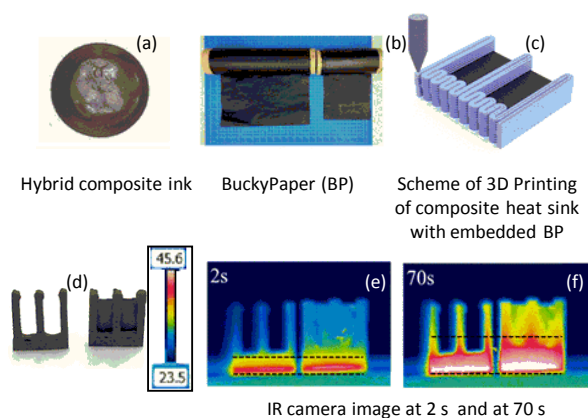

Fig. 5. (a - c) Schematic of buckypaper heat sink. (a) Composite ink obtained by epoxy filled with graphite-nanoplatelets and carbon fiber. (b) Free standing carbon nanotube films, or buckypaper (BP). (c) Printed heat sink using composite ink, with BP incorporated. (d) Heat sinks without (on the left) and with BP (on the right). (e - f) Heat dissipation as mesured from IR camera of two heat sinks at different time frames. Reprinted with permission from [58]. Copyright 2021 American Chemical Society. Original work has been modified with permission.
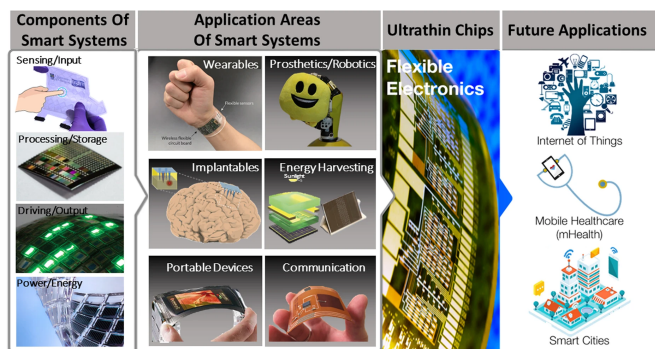

Fig. 6. An overview of flexible devices. Original work from [59], reproduced under CC-BY-4.0.

Commonly, an enhanced heat dissipation is achieved by increasing the heat exchange rate with the environment through a heat sink connected to the heated structure. However, solid surfaces are rough, and this introduces extremely thermally insulating air gaps in between, hindering the heat exchange. To mitigate this issue, TIMs are used at the junctions of different materials to fill these gaps. TIMs are required to possess 1) excellent thermal conductivity, and 2) suitable Young's modulus. In this respect, CNTs, graphene and related composites are widely employed as TIMs.

CNTs possess high thermal conductivity, together with low transverse elastic modulus, the latter making these able to support external loads without permanent deformation. Different factors, such as orientation, density, aspect ratio, structure, and defects have proven to affect the thermal conductivity of CNTs [60]. Vertical arrays of CNTs (VACNTs) are ideally suited as TIMs, as they show better thermal conductivity compared to mats of randomly oriented CNTs. Also, they have good mechanical compliance, and thus they are able to absorb the stress induced by the mismatched coefficient of thermal expansion of a device, resulting in lower interfacial

thermal resistance [61]. By increasing the CNT density or by metal bonding, VACNTs have demonstrated to improve both the thermal conductivity and the contact thermal resistance [62], [63]. Additionally, they show good resilience to thermal cycling, which guarantees reliability under operation conditions. Polymer-based TIMs with CNTs as fillers may combine the high thermal conductivity of CNT with the polymer compliance. Their thermal conductivity is strongly influenced by the polymer, and by the quality, density and orientation of CNTs. Free standing carbon nanotube films (buckypaper) with thermal conductivity of 20 W/mK and large surface area have been mass produced in-house and have shown to be a lightweight solution for thermal management devices [58].

Being 1D materials, CNTs possess high thermal conductivity only along the axis, whereas graphene can transfer heat along the plane. However, the thermal conductivity of graphene was found to be anisotropic between the in-plane and the out-of-plane orientations. As previously explained, this is due to the strong covalent sp^2 bonds of in-plane C atoms and the weaker van der Waals interaction between graphene monolayers. Various startegies have been adopted to enhance the out-of-plane thermal conductivity of graphene, which has important implications for the heat transfer from device to heat sink. For instance, the fabrication of a graphene-based microstructure, with vertical graphene layers sandwiched between horizontal graphene layers, has achieved a compressive module as low as 0.87 MPa and an out-of-plane thermal conductivity of 143 W/mK, higher than those of many metals [64]. The use of graphene as a filler has demonstrated to increase the thermal conductivity of polymer-based TIMs much more than employing CNTs with an equal filler fraction [65].

3D-carbon-based materials, like graphene foams and graphene aerogels, are excellent TIM candidates [36]. Their intrinsic high porosity lowers the thermal conductivity, which however is isotropic. Moreover, they show high flexibility under compression, and they are free standing. Graphene foams are synthesized growing graphene on Ni or polyurethane foams, and then by etching the template. The porosity of such foams can be reduced by prolonging the growth time, or by compression, and this increases their thermal conductivity. Graphene aerogels possess smaller pore size compared to foams, leading to a higher number of thermally conductive paths, whereas their excellent elasticity allows to maintain the structure under compression. Similarly to graphene foams, the thermal conductivity of aerogels can be increased making them denser by compression.

C. Flexible Electronics

The possibility to fabricate flexible devices built on stretchable substrates is attracting a great deal of regard from both academia and industry. An impressive number of application areas is envisioned in the near future for such devices: few notable examples are healthcare (pain relief patches, wearable sensors to monitor several health parameters), well-being (wearables for sport), smart buildings (sensors for temperature and moisture level), photovoltaics (flexible solar cells for

indoor and outdoor activities) and other energy harvesting devices (flexible batteries and capacitors), and automotive industry (displays, sensors). Nonetheless, the main drive to the growth of flexible electronics market is the development of smart and connected devices: flexible radio frequency identification (RFID) tags and sensors are considered to be the technological bricks of the Internet of Things (IoT).

Recently, the development of the Internet of Medical Things (IoMT), made of wearable devices and sensors to provide real-time patient/doctor interaction, has attracted huge interest. Different types of wearable sensors will be necessary to build the IoMT ecosystem, such as mechanical, optical, biochemical, and biopotential sensors to monitor several health parameters [66]. Possible candidates as sensing layers in flexible devices are CNT-based nanocomposites, where CNTs are integrated into the polymer matrix as conductive fillers. The working mechanism of CNT/polymer nanocomposites can be explained by means of the percolation theory. The network resistance depends on various factors, such as the tunneling resistance at CNT junctions, and the morphology of the CNT network. By streching the polymer matrix, the CNT network is altered, with the subsequent breaking and formation of CNT junctions. Overall, as it is expected that the number of broken junctions increases with tensile strain compared to the number of newly formed junctions, the network resistivity is essentially dependent on the tensile strain. Thus, CNT/polymer nanocomposites are ideally suited for wearable health monitoring mechanical sensors [31], [32].

IV. CONCLUSION

In this work, we have reviewed the recent progress in electronics nanopackaging based on CNTs, graphene, and related materials. In particular, now that the Moore's law is approaching its physical bottleneck, an analogue law for packaging [37] could, by system scaling, continue to push the efficiency improvement of ICT in the next years. First, we have illustrated the relevant properties of graphene and CNTs for applications in electrical packaging, that is their electrical, thermal, and mechanical characteristics. Then, we have described the enormous asset represented by graphene and CNTs to overcome the issues correlated to the trend of device downscaling for: 1) interconnects, and 2) thermal management, two of the main areas of interest in nanopackaging. Finally, we have also briefly illustrated the application of CNTs in flexible electronics, this latter being on the forefront for the development of IoT.

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