Natural Graphite Sheet Heat Sinks:
A Review of the Material Properties, Benefits, and Challenges

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Abstract

A multidisciplinary feasibility study of natural graphite sheet (NGS) heat sinks, addressing the thermal, EMI/EMC, reliability, cost and manufacturing perspectives, is presented together with a brief overview of the relevant graphitic materials and a summary of the NGS properties (thermal conductivity, heat capacity, weight, electrical conductivity, emissivity, coefficient of thermal expansion). The high in-plane thermal conductivity (up to 600 $W \cdot m^{-1} \cdot K^{-1}$) of NGS and low interface resistance of NGS heat sinks can lead to good thermal performance despite the low through-plane thermal conductivity of NGS, but new designs are required. Low electrical conductivity offers a potential to decrease conducted and radiated electromagnetic emissions and significant weight reduction can be achieved due to the low density of NGS. The raw material cost of natural graphite is shown to be lower than aluminum and a high-volume manufacturing method is discussed. Low mechanical strength of NGS does not allow mounting using threaded holes in the heat sink, limits applications where a chance collision with other objects exists, and might have an impact on reliability of large power modules.

Keywords

Heat sinks, natural graphite, flake graphite, compressed expanded natural graphite, kish graphite, material properties, electromagnetic interference, EMI, electromagnetic compliance, EMC, reliability.

Nomenclature

NGS – Natural graphite sheet
TIM – Thermal interface material

1. Introduction

In our proof-of-concept study [1] we manufactured a prototype natural graphite sheet (NGS) heat sink and showed that its thermal performance is equal to or better than a geometrically identical aluminum heat sink. This paper aims to give a review of the graphitic materials relevant to thermal management, the properties of NGS, and further investigate the potential of NGS heat sinks from a multi-disciplinary perspective. Although heat sinks are often evaluated solely based on thermal performance, electromagnetic performance, reliability and cost of electronic devices are also important. For an emerging heat sink material, it is crucial to evaluate all aspects to target the right applications and to avoid complications related to switching from a conventional to a new material.

2. Review of graphite

Graphite is a form of carbon that consists of stacked layers of graphene with the characteristic hexagonal arrangement of atoms. Atoms within each layer are bonded by a strong covalent bond, while the layers are kept together by weak van der Walls forces. It is common to denote the directions in the plane of graphite as a and b, and the perpendicular direction as c. The difference in the atomic bonds in the ab and c directions translates into the anisotropy of graphite properties such that, for example, the thermal conductivity is estimated to be 2000 $W \cdot m^{-1} \cdot K^{-1}$ in the ab-directions, and 6-9 $W \cdot m^{-1} \cdot K^{-1}$ in the c-direction [2]. The staggeringly high thermal conductivity attracts a significant amount of attention, but it is important to note that the numbers relate to a monolithic (single crystal) structure with no defects. In reality, all graphite materials are polycrystalline, i.e. they consist of many crystals whose size and orientation are characteristic for each of the materials. Larger crystals oriented in the same direction result in materials with properties closer to the single crystal.

Pyrolytic Graphite Sheet (PGS) is a thin graphitic material whose structure consists of large crystals oriented perpendicular to the thickness. It has become very popular in the electronics cooling industry for its good spreading performance arising from an in-plane thermal conductivity of up to 1950 $W \cdot m^{-1} \cdot K^{-1}$ [3][4]. PGS is manufactured from polyimide films such as Kapton in a process that requires reaching graphitization temperatures of approximately 3000 °C [5].

NGS is manufactured from flakes, which are found trapped in soft or hard host rock in deposits around the world. Crushing, milling and flotation yield free flakes, which, however, typically need further chemical and/or thermal treatment to eliminate impurities such as sulphur or iron [6]. Purified flakes (Figure 1a) are treated with an intercalation agent (most often sulfuric acid), which penetrates in between the graphite layers. This so-called graphite intercalation compound or expandable graphite is then rapidly heated which forces the intercalation agent to expand and push the individual carbon layers apart, forming exfoliated graphite, known for its worm-like or accordion-like structure (Figure 1b). Compressing the exfoliated graphite yields cohesive sheets whose thickness range from hundreds of microns to several millimetres (Figure 1c). The degree of orientation of the graphite layers increases with the level of compression, and thus the properties of the sheet change as well. Multiple names are used for these sheets - flexible graphite sheets, compressed exfoliated graphite, or NGS. The first name, which relates to the through plane compressibility of the original application as seals and gaskets, is misleading in the context of this paper as the thick sheets that

Figure 1: Manufacturing of Natural Graphite Sheet (NGS). a) flake graphite before exfoliation, b) after exfoliation, c) compressed exfoliated graphite. (Photographs reprinted from [7] with a permission from George Simandl)

we used are not flexible when compared to PGS as shown in Figure 2. Throughout this text, the term NGS is used, but Compressed Exfoliated/Expanded Natural Graphite (CENG) is equally appropriate.

Two other distinctive types of graphite exist – synthetic and kish. The former is manufactured from petroleum coke and coal tar pitch in a process that involves heat treatment at high temperatures (up to 3000°C [8]) and forming by extrusion or pressing of simple shapes whose properties are fairly isotropic due to the random orientation of the crystals. Relatively low thermal conductivity of 70 to 140 $W \cdot m^{-1} \cdot K^{-1}$ [9] in combination with the need for costly machining to form complex shapes does not make synthetic graphite an attractive material for heat sinks.

Kish graphite has very similar structure and properties to the natural flake graphite, but instead of mining, it is obtained as a by-product from the steelmaking process. Steel is an alloy containing mainly iron and carbon. At certain temperatures and concentrations, the molten alloy becomes supersaturated with carbon, which leads to its segregation into flakes that are generally bigger than natural graphite flakes and also contain less defects in the crystal structure [10],[9]. Despite its similarity to natural graphite, to the best knowledge of the authors, kish graphite sheets are not commercially available and only one publication [11] demonstrates the possibility of exfoliating kish graphite and compressing it into sheets, but it does not involve measurement of thermal properties. Small batches of kish graphite are commercially available and are typically used for preparing graphene (a single layer of graphite) for scientific studies. Whether the high price (30 USD per gram [12]) is a result of low competition in the niche market, or a valid indicator of the production cost is not known to the author.

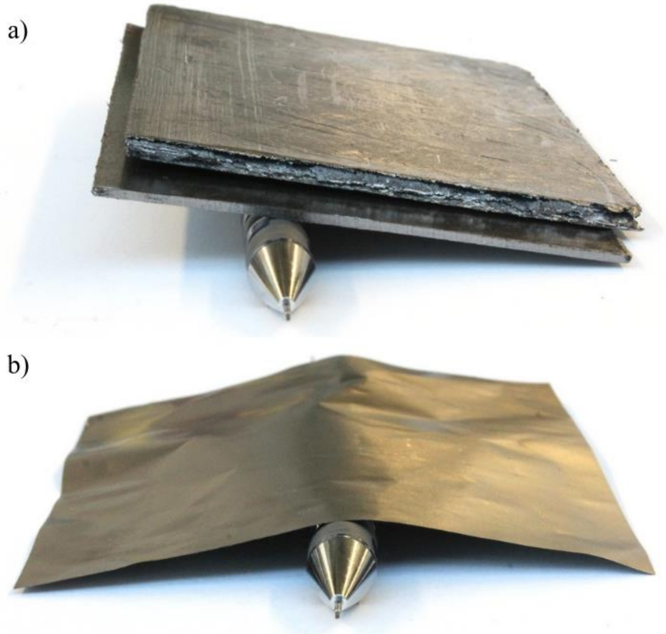

Figure 2: Illustration of flexibility: a) Natural Graphite Sheet (NGS) b) Pyrolytic Graphite Sheet (PGS)

3. Properties of NGS

The properties of NGS are dependent on the size and chemical purity of the flakes and, most of all, on the level of compression, typically expressed in terms of density. A simplified graphical summary of the properties averaged from [13]–[20] is shown in Figure 3.

While the in-plane thermal conductivity increases with density, the through-plane thermal conductivity shows the opposite trend, i.e. it decreases with density. The specific heat capacity, which increases with temperature, was reported to be in the range of 711-850 $Jkg^{-1}K^{-1}$ [18][13][14]. The lower values seem to relate to the room temperature, while the higher values may be averages over a certain temperature range, but a definitive conclusion cannot be made due to the limited information in the literature sources. The In-plane coefficient of thermal expansion is very small and negative (-0.4 [$10^{-6} \cdot K^{-1}$]

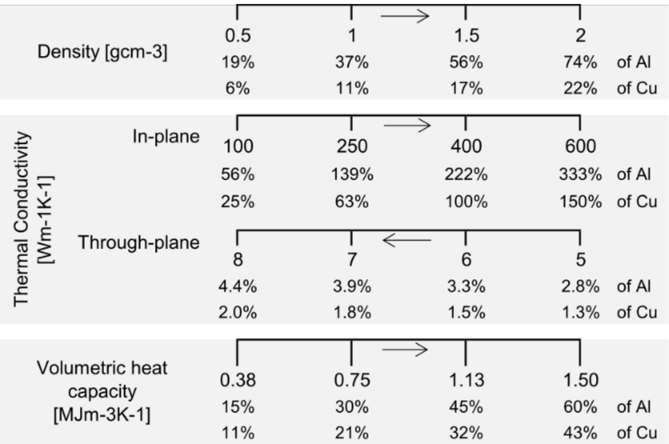

Figure 3: Approximate properties of natural graphite sheet (NGS). Reference values: ρ_{Al}=2.7 $g \cdot cm^{-3}$, ρ_{Cu}=8.9 $g \cdot cm^{-3}$, k_{Al}=180 $W \cdot m^{-1} \cdot K^{-1}$, k_{Cu}= 400 $W \cdot m^{-1} \cdot K^{-1}$, $c_{P,Al}$=910 $J \cdot kg^{-1} \cdot K^{-1}$, $c_{P,Cu}$= 390 $J \cdot kg^{-1} \cdot K^{-1}$, $c_{P,Gr}$= 750 $J \cdot kg^{-1} \cdot K^{-1}$. Data: [12]-[19].

[21] [22]), which means that in the in-plane direction NGS shrinks with increasing temperature, but the through-plane coefficient of thermal expansion is relatively high ($27[10^{-6}\cdot K^{-1}]$ [21][22]) and comparable to aluminum. The density dependence of the expansion coefficient has not been reported. Emissivity of NGS is 0.43 at 816 °C [18] and 0.4 at 400 °C [21], but the data for room and low temperatures seem to not be published. Analogously to the thermal conductivity, the electrical conductivity is higher and increases with density in the in-plane direction, while in the through-plane direction it is lower and decreases with density. Whether NGS follows the Wiedemann–Franz law, which relates the electrical and thermal conductivities, is questionable as opposing conclusions can be found in the literature. Graphite has been reported to be diamagnetic [23].

The material cost was estimated to be 2.2 USD per dm^3 for natural graphite and 4.8 USD per dm^3 for aluminum based on [24], assuming the densities to be 1.6 $g\cdot cm^{-3}$ and 2.7 $g\cdot cm^{-3}$, respectively. The price per volume was chosen to allow a simple comparison of two geometrically identical products.

Multiple resources ([25]–[29]) were used to compile Figure **4**, which shows the volumetric GER (gross energy requirement) for graphite and conventional heat sink materials. Even though the data for both natural and synthetic graphite is related to anodes for lithium ion batteries, it is possible to conclude that production of one cubic meter of natural graphite requires less energy than the other materials in the comparison. Other factors such as water requirements, toxic waste production, and emissions to the atmosphere also influence the overall environmental footprint of graphite production. As the biggest exporters are often countries with poor reinforcement of environmental regulations, the environmental footprint is usually overlooked to drive the price down. Graphite can be sourced ethically by carefully choosing the supplier.

4. NGS heat sinks

4.1. Thermal performance

NGS heat sinks are manufactured by cutting the graphite sheets into the desired shape and gluing them together in a stack as discussed in [1]. An example of a staggered plate fin NGS heat sink is shown in Figure 5, but a variety of other shapes and sizes can be produced using this simple method.

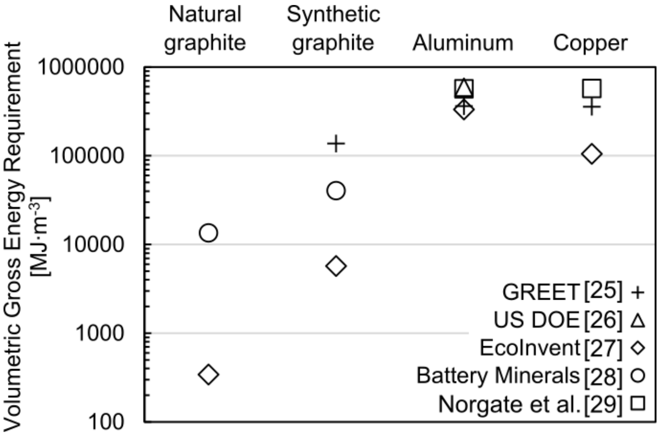

Figure 4: Volumetric gross energy requirements of graphite, aluminum, and copper. The vertical axis is logarithmic.

Figure 5: Staggered plate fin NGS heat sink

NGS heat sinks inherit the anisotropy of the sheets, i.e. the heat conduction in one direction is significantly lower than in the other two, which limits the heat spreading in the base plate as reported in [30].Figure 6a) and b) show aluminum and NGS heat sinks mounted on small heat sources and illustrate that while the heat spreads uniformly into all the fins of the aluminum heat sink, the spreading in the base plate of the NGS heat sinks is limited by the low thermal conductivity in the x-direction, which negatively affects the overall thermal performance. To achieve the best performance the width of a NGS heat sink should be matched to the width of the heat source, which eliminates the base spreading problem, but limits the number of fins and thus requires taller fins to compensate for the lost heat transfer area as shown in Figure 6c. Taller fins reduce the fin efficiency, but if an NGS is selected with a thermal conductivity in the y-direction (in-plane) that is higher than aluminum, increasing the fin height is feasible as illustrated by the following simple 2D case study. To replace the aluminum heat sink outlined in Figure 6a (number of fins: 6, fin height: 20 mm, fin thickness: 0.5 mm, thermal conductivity: 200 $W\cdot m^{-1}\cdot K^{-1}$, convective heat transfer coefficient: 30 $W\cdot m^{-2}\cdot K^{-1}$, fin efficiency 92.7 %) with an

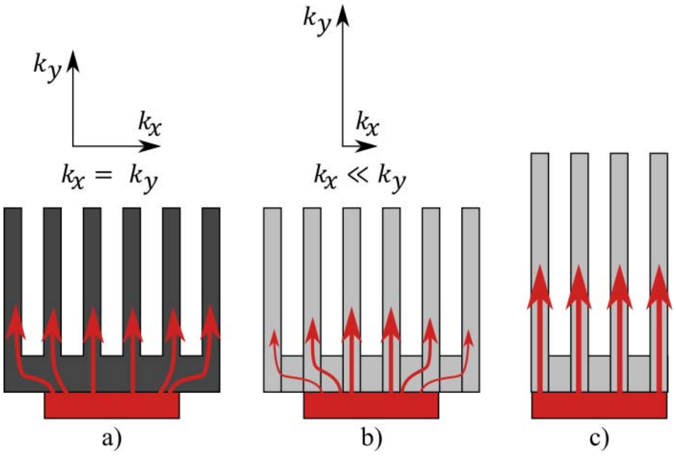

Figure 6: Heat spreading in the base plate of a NGS heat sink. a) Isotropic metal with thermal conductivities equal in both directions $k_x = k_y$, b) anisotropic graphite heat sink with k_x ≪ k_y, c) modified graphite heat sink design to eliminate the spreading problem

equally performing one made of NGS, but with four fins (Figure **6**c), the fin height needs to increase to 30 mm, which causes only a negligible decrease in fin efficiency (to 91.7 %) as the thermal conductivity of NGS is higher (400 $W\cdot m^{-1}\cdot K^{-1}$). The weight of the NGS heat sink is 56 % of the aluminum one (assuming the density of aluminum and NGS to be 2.7 $g\cdot cm^{-3}$ and 1.5 $g\cdot cm^{-3}$, respectively). While the example above illustrates the basic principle, finding a matching NGS heat sink to a general aluminum heat sink is case dependent because many variables are involved (geometry, thermal conductivity, convective heat transfer coefficient). It is likely that in cases of wide, low profile heat sinks on small heat sources the replacement with a NGS one is not possible even if very high density NGS with thermal conductivity of 600 $W\cdot m^{-1}\cdot K^{-1}$ is used.

A very attractive property of NGS heat sinks is the conformity in contact with hard surfaces. Commercially available graphite thermal interface materials utilize this property to reduce the thermal contact resistance between heat sinks and the base plates of power modules. Graphite conformity, which arises from its flake structure and significantly lower hardness, is beneficial on two scales – on the macroscopic scale a NGS heat sink will deform and conform to out-of-flatness features as shown in Figure 7c; on the microscopic level the flakes will conform to the micro roughness, which is to some extent present on all engineering surfaces (Figure 7d). Metal heat sinks will not conform either on the macro (Figure 7a) or microscopic scale (Figure 7b) and the heat is forced to constrict and flow only through a fraction of the contact area, resulting in high contact resistance, which is commonly mitigated by filling the voids with a thermal interface material. Our recent measurements [31] suggest that at 0.5 MPa clamping pressure the interface resistance of a NGS heat sink is comparable to an aluminum one with thermal grease, which can have immense implications on reliability as discussed in section 4.3.

4.2. Electromagnetic interference and compliance

Heat sinks affect the performance of electronic devices and can become the reason for failing the Electromagnetic Compatibility (EMC) tests [32][33]. To demonstrate the commonly encountered problems in a practical manner, we discuss several typical ways to cool a simple discrete power semiconductor and their implications on the thermal and electrical performance of the device. In most discrete semiconductor packages, the exposed metal part, known as the lead frame, tab, or heat slug, is electrically connected to one of the terminals (cathode in diodes, source in MOSFET transistors), which means that whatever voltage exists on the terminal is also present on the lead frame. The simplest cooling solution, which is very attractive thermally, is attaching the device directly to the heat sink without any electrical insulation, making the heat sink electrically live (Figure 8). The heat sink follows the voltage of the cathode or source and, if the device operates at high frequencies, it becomes a voltage driven antenna, whose radiated power depends on the geometry and the material properties of the heat sink. In radio communication, where the goal is exactly opposite that in heat sinks, i.e. maximizing the radiated power of an antenna, the dimensions are chosen so the antenna resonates at the design frequency, e.g. the length of a monopole antenna is a quarter of the wavelength. In heat sinks, the goal is to avoid resonance, but predicting the resonant frequencies is a complicated task that typically requires numerical simulations in specialized finite element electromagnetics solvers. A general rule to evaluate the potential risk of radiated emissions is to check if the heat sink size and the wavelength are comparable. For example, a 12-cm heat sink is prone to resonating at frequencies around 2.4GHz, and smaller heat sinks resonate at higher frequencies. While integrated chips can operate at such high frequencies, the kHz to MHz switching frequencies of power electronics may seem too low to cause significant radiation, but the rapid changes in voltage and current (dV/dt and dI/dt) during the switching result in a high frequency content of the voltage signal that can reach the critical gigahertz spectrum.

In most practical applications where touch safety is required and where multiple devices are mounted on the same heat sink, electrically insulating thermal interface materials are used to isolate the heat sink, which generally reduces the radiated emissions, especially if the heat sink is grounded [34] [35], but the interface thermal resistance increases significantly. At the

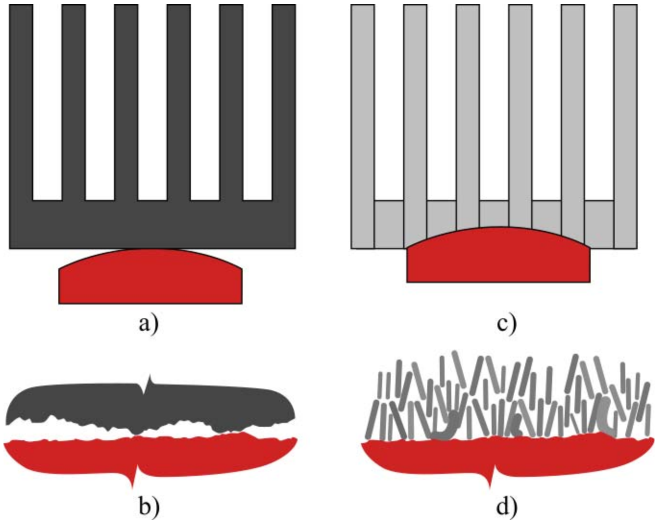
a) c)
b) d)

Figure 7: Illustration of the interface conformity: a) graphite macro-scale, b) graphite micro-scale, c) aluminum macro-scale, d) aluminum micro-scale

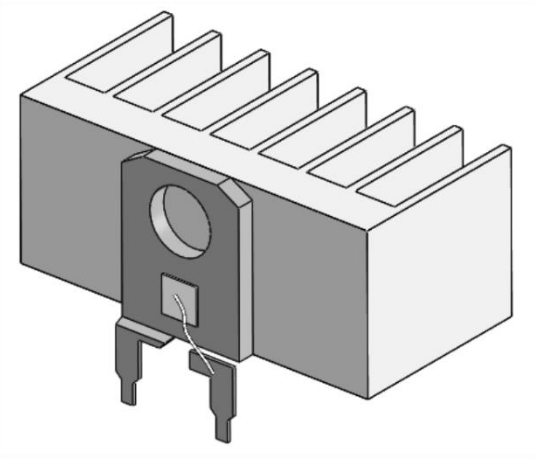

Figure 8: Power semiconductor mounted on a heat sink without electrical insulation (a TO-247 diode is shown without the molding compound to make the internal structure visible)

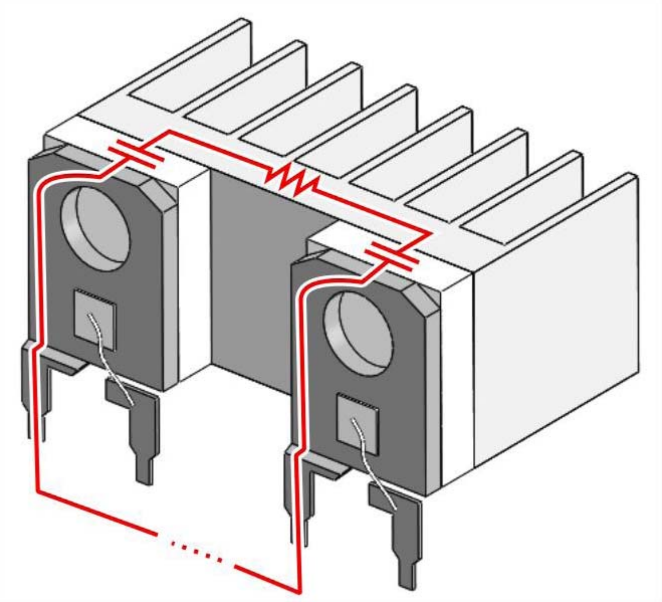

Figure 9: Two semiconductors mounted on a same heat sink with electrical insulation. The equivalent circuit for parasitic currents (conducted emissions) is shown in red.

same time a capacitor is formed at the interface whose charging and discharging degrades the switching performance of the system, and through which parasitic currents can flow as shown in Figure 9. The parasitic currents are often referred to as conducted emissions. The equivalent circuit in Figure 9 includes the impedance of the heat sink, which is usually neglected due to the low electrical resistivity of aluminum.

As shown in Figure 10, the electrical resistivity of NGS is orders-of-magnitude higher than aluminum or copper, which suggests that significant reductions in both the radiated and conducted emissions can be achieved. The mechanism of the reduction is a simple increase in impedance, which leads to lower currents. In case of radiated emissions, it is assumed that a part of the energy that would be radiated from a high-electrical-conductivity heat sink will be dissipated as heat in a low-electrical-conductivity heat sink.

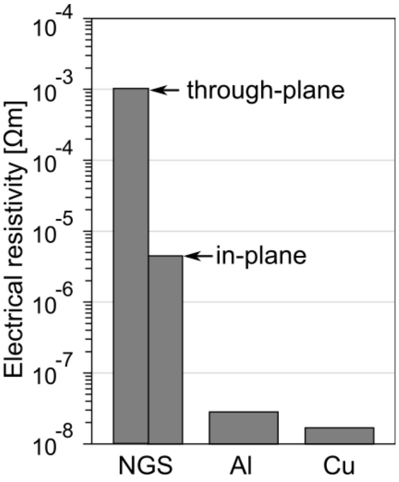

Figure 10: Comparison of the electrical resistivity of NGS, aluminum, and copper. The vertical axis is logarithmic. NGS data from [19][13], Al and Cu data are approximate.

The main implications for power electronics are that a simple change in the heat sink material can decrease the emissions and reduce the size of the EMI filter circuitry, and that in cases where using aluminum heat sinks without electrical insulation was rejected due to high radiated emissions, NGS heat sinks could be feasible.

In addition to reducing the emitted radiated power, NGS heat sinks also absorb incoming electromagnetic waves instead of reflecting them as metals [17], which can lead to decreased levels of electromagnetic fields within the chassis of electronic devices.

4.3. Reliability

Reliability of power electronics is closely related to thermal management, mainly due to thermomechanical stresses that arise from temperature cycling. Before discussing the degradation mechanisms in detail, one obvious advantage of graphite based cooling systems can be demonstrated based on the findings presented in the previous sections. As graphite heat sinks do not require thermal interface material, a major concern in long term reliability is eliminated. Paste-like and phase change TIMs require careful application to assure the right amount and distribution and their performance over the life time of a device is a concern due to the pump out, dry out, and/or chemical degradation. Increased operating temperatures of new semiconductor materials, such as Silicon Carbide (SiC), intensify the degradation process of common TIM’s, however, graphite is stable up to approximately 400 °C [21] [20].

Materials used in semiconductor packaging (copper, aluminum, ceramics, solder, etc.) have different coefficients of thermal expansion (CTE), which, together with non-uniform temperature distributions across devices, result in thermomechanical stresses at the interfaces where the materials are in contact [36]. Larger temperature gradients and CTE mismatches result in higher stresses. Real life electronic devices do not operate at steady temperatures, instead they undergo cyclic loading due to time-varying power and ambient conditions, which results in crack propagation and degradation of electrical connections.

The reliability implications of using NGS heat sinks can be qualitatively estimated by assuming a system in which an aluminum heat sink was replaced by an NGS one. It is convenient to split the discussion in two parts: purely thermal and thermomechanical.

Thermally, the main difference is the transient performance in fluctuating load. Since the volumetric heat capacity of NGS is much lower than of aluminum (Figure 3), the temperature within the device will change more rapidly, which could threaten reliability. It is, however, important to also consider the thermal resistance of the heat sinks. If the NGS heat sink offers a lower thermal resistance, the maximum temperature of the device will decrease, which should have a positive effect on reliability.

From the thermomechanical perspective, the main factor is assumed to be lower stiffness of the NGS heat sink, which will affect the deformation and stresses within the device. Studies of power semiconductor reliability tend to focus solely on the packaging and neglect the influence of heat sinks, which might be reasonable for small discrete devices or power modules mounted on large stiff aluminum heat sinks, but is not appropriate in the case of large modules on NGS heat sinks. Chapter 11.6.6. in [36] states that mounting a power module on a heat sink reduces the bimetallic flexing and extends the lifetime. The lower stiffness of NGS heatsink could allow for increased flexing and thus lead to lower lifetimes. However, due to the large variation in power modules design and the complexity of the topic, this should be perceived as a potential issue.

While the conformity of NGS was shown to be an advantage as thermal greases are no longer necessary, the long-term degradation of the graphite-metal interface has not been studied and most certainly requires attention.

High chemical stability of graphite suggests its feasibility for applications in harsh environments. Ability of synthetic graphite to withstand acidic environment has been shown in [37]. Whether the results can be extrapolated to NGS is not guaranteed, but the fact that acids are used in the manufacturing of NGS supports this assumption. Oxidation of natural graphite in air occurs only at high temperatures (600 °C) and its rate is dependent on multiple parameters such as the flake size or the amount of impurities [38]. Assessing the corrosion behavior in humid or even marine conditions requires a complex electrochemical analysis and/or experimental studies. Since graphite is more noble than copper or tin, which are typically used for the surfaces of electronic devices that are adjacent to a heat sink, a risk of galvanic corrosion resulting in damage of the device exists [39], but the relevant measurements have not been reported in the literature.

4.4. Manufacturing and cost

To the best knowledge of the authors, NGS heat sinks are not currently produced on a commercial scale, and thus the manufacturing cost can be estimated only qualitatively. While virtually all aluminum and copper forming methods require heavy machinery and often also high temperatures, the equipment for shaping soft graphite and assembling the sheets into final laminates is fairly simple. This is expected to lead to lower capital and operating costs. Production of NGS parts has a long history in the fuel cell industry and bipolar plates are typically produced using roll embossing technology at a throughput of 9 metres per minute. The technology is expected to be easily transferrable as heat sink shapes are significantly less complex in comparison with bipolar plates, which include delicate features such as flow channels as narrow as 800 μm.

4.5. Challenges

Low mechanical strength makes the NGS heat sinks prone to breaking when impacted by other objects in applications where a risk of a collision exists. While methods to improve the mechanical strength of NGS exist (resin impregnation), the author believes that true potential of NGS heat sinks is in devices that are fully protected by a mechanically sound casing.

Another challenge arising from the low mechanical strength is mounting using bolts. Making a threaded hole in the base of a NGS heat sink is not possible and other clamping methods are necessary. If weight savings are the main motivation in employing NGS heat sinks, a poor clamping design using too much material could reduce or even completely eliminate the weight advantage of NGS.

Operation in a vibrating environment could lead to detachment of flakes from the NGS structure, creating a risk of short circuiting exposed terminal within the device. While no studies to prove or refute this concern have been reported, the same issue has been addressed in the fuel cell industry by impregnating the NGS with resin. In the context of NGS heat sinks, the resin treatment is relevant, but in selecting the resin its effect on the thermal contact resistance, maximum operating temperature and other critical properties must be considered.

5. Conclusion

An extensive multidisciplinary overview of NGS heat sinks was presented. NGS is a graphitic material whose properties are anisotropic, density dependent, and in many cases the properties are not available and further measurements are required. High in-plane thermal conductivity makes NGS a good candidate for a heat sink material despite the low through-plane thermal conductivity, which can be mitigated by an appropriate design of the heat sink. The low hardness of NGS results in low thermal contact resistance that is comparable to metal heat sinks with thermal grease, and which offers a possibility to eliminate the grease from the cooling assemblies and increase the overall reliability. On the other hand, the lower stiffness of NGS heat sinks may negatively affect the device reliability by allowing bimetallic bending deformation. Comprehensive reliability studies are required to answer these complicated questions. From the electromagnetic interference perspective, NGS heat sinks have the potential to reduce both the conducted and radiated emissions as their high electric resistivity can damp the parasitic currents. The low cost of the material and the simplicity of the manufacturing process suggest that NGS heat sinks can be economically competitive. Graphite requires the lowest amount of energy per production of one cubic meter in comparison with aluminum and copper, which potentially makes it an environmentally conscious choice, but other aspects of the production, such as acid use, must be reviewed. Due to the low mechanical strength of NGS, mounting using a threaded hole in the heat sink is not feasible, and it is not recommended for applications where a chance of collision with other objects exists.

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