Investigation of Hybrid Graphene-hBN and Graphene-GO as a Direct Contact Heat Spreader

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Abstract—This article studies the suitability of hybrid graphene-hexagonal boron nitride (G-hBN) and graphene-graphene oxide (G-GO) as direct contact heat spreaders on a Pt/Cu/Ti micro-coil. The Ar/CH₄ plasma treatment converts hBN and GO into G-hBN and G-GO heat spreaders, respectively based on the appearance of graphene Raman I(G) peak at ~1575 cm⁻¹. Both G-hBN and G-GO are superior heat spreaders compared to the initial hBN and GO with more than 0.5-times of hotspot temperature reduction, 10-times more coil cooling effects, and more than 0.5-times reduction in lateral thermal resistance. However, the plasma treatment is damaging to the hotspot device and causes a severe increase in the coil resistance of at least 1.2-times due to thinning of metal. Therefore, the preferential heat spreader is still the non-hybridized hBN since it negligibly affects the resistance of the Pt/Cu/Ti coil. It offers a ~16% reduction in hotspot temperature and reduces the lateral thermal resistance by ~37.4%. It is the third-best after the G-GO and G-hBN heat spreaders.

Keywords—heat spreader, 2D insulator, graphene-hBN, graphene-GO

I. INTRODUCTION

Employing the high thermal conductivity, \( k \) of two-dimensional (2D) materials as a lateral heat spreader may provide a robust solution for alleviating thermal degradation in power devices [1]. One of the techniques for thermal management is by incorporating a 2D heat spreader to carry heat from the hotspot to the nearest heat sinks. For this purpose, 2D materials with high values of \( k \), e.g. graphene with \( k>1600 \text{ Wm}^{-1}\text{K}^{-1} \) [2] and hexagonal boron nitride (hBN) with \( k>750 \text{ Wm}^{-1}\text{K}^{-1} \) [3] are both promising. In recent works, we introduced the use of Pt/Cu/Ti thermal test structure to study the lateral heat spreading of multilayer graphene [4] and vertical graphene [5] during steady- and transient-state Joule heating. Unfortunately, direct contact of conductive graphene on the metal spreads the current and rendered our resistance thermometer unusable. Gao et al. [6] and Zhang et al. [7] reported the use of Au/Pt/Ti micro-coil heater-thermometer to study the performance of graphene heat spreaders. To avoid short-circuiting the coil, they deposited a thin SiO₂ on the coil before the deposition of graphene. Although the system is capable to demonstrate the function of graphene as a lateral heat spreader, the thin SiO₂ interlayer contradicts as it introduced an additional blockage within the critical heat path. Unlike graphene, the use of insulator 2D materials, i.e. hBN and graphene oxide (GO) allows direct deposition on hotspots without short-circuiting the device. In this article, we investigated the performance of hBN and GO as 2D direct contact insulating heat spreaders. Then, both materials were hybridized with graphene into graphene-hBN (G-hBN) and graphene-GO (G-GO) films as a 2D semi-insulator. The comparison between single and hybridized films was carried out to conclude the suitability of the modified hBN and GO as direct contact heat spreaders.

II. EXPERIMENTAL

The thermal test structure consists of a central coil and peripheral heat sinks were fabricated using standard lithography and Pt/Cu/Ti metallization process on SiO₂/Si substrate. Multilayer hBN film was grown on Ni foil by a thermal CVD system (Oxford Instruments Nanofab 1000) at 1050 °C using B₂H₆ and NH₃ precursors. The hBN heat spreader was formed by wet transfer of hBN using polymethyl methacrylate carrier onto a thermal test structure. The GO heat spreader was obtained by spin-coating of 1 mg ml⁻¹ commercial GO (Graphene Supermarket) onto a thermal test structure. Both samples of hBN- and GO-coated coil were

![Fig. 1](image-url)
dried on a hotplate at 100 °C for 10 min before the electrothermal characterization. Using the same samples, hBN and GO films were converted into G-hBN and G-GO films by Ar/CH₄ plasma at 600 °C as elaborated elsewhere [5].

The thickness of as-deposited hBN and GO films were measured using the fixed angle of 75° spectroscopic ellipsometry technique with a DUV-UV-visible detector (Semilab SE-2000). The fingerprints of hBN, G-hBN, GO, and G-GO were studied using λ = 514 nm confocal Raman spectroscopy system (WITec alpha300R). The electrical measurement was done using a four-wire Kelvin configuration (NEXTRON mini probe station), a source meter (Keithley 2410), and an IR thermal imaging camera (FLIR ETS320). Details on electrothermal characterization setup using Pt/Cu/Ti were elaborated elsewhere [8].

III. RESULTS AND DISCUSSION

The thickness of as-deposited hBN and GO films were measured using a three-phase structure of ‘air/hBN/void/SiO₂/substrate’, while GO was using a two-phase structure of ‘air/GO/SiO₂/substrate’. The optical model was using Kramers-Kronig consistent Tauc-Lorentz dispersion law [9]. It results in a satisfactory good fit on the ellipsometry responses with R² above 0.99. The thickness of wet transferred hBN was 11.5 ± 0.8 nm, while the thickness of the spin-coated GO film was 14.1 ± 0.6 nm. Fig. 1a shows the photo of the thermal test structure. The central coil serves both as a heat source and resistance thermometer, while the peripheral metals serve as heat sinks. The hotspot generation is at the center of the coil as indicated in Fig. 1b using locked-in thermal imaging. The equivalent thermal resistance, Θ network is shown in Fig. 1c, where the generated heat from the coil Joule heating power, Pcoil travels laterally and vertically to the sinks. The hotspot temperature read by the resistance thermometer is denoted by T₀, while the surface resistance T₁ and T₂ were read by the IR thermometer. All these three temperatures are essential for analyzing the performance of 2D insulator lateral heat spreaders. The deposition of 2D materials on the coil will modify the Θ network by providing a shunt for the lateral heat path, from the coil to the peripheral heat sinks.

Fig. 2a is the photo of the transferred hBN on the coil. The hBN film was larger than 7×7 mm² to carry the heat from the 5×5 mm² central coil to the peripheral heat sinks. Thermograph in Fig. 2b shows the hBN-coated coil at an applied bias of 20 V and it indicates the respective temperature points. Raman spectra and surface condition of transferred hBN on the coil are provided in Fig. 2c and 2d, respectively. The weak E₂g peak was found at ~1368 cm⁻¹, verifying an hBN fingerprint [8]. The contrast of multilayer hBN was noticeable from the optical microscopy image. Fig. 2e and 2f are the Raman spectra and surface condition of the G-hBN-coated coil, respectively. After the Ar/CH₄ plasma treatment, defect peak intensity was increased with I(D)/I(G) of 1.277. In this case, G-GO is simply a highly reduced GO by the Ar/CH₄ plasma treatment. The G-GO film appeared darker than the original GO film.

The temperature-power (T-P) profile in terms of T₀, T₁, and T₂ for the hBN and G-hBN characterization is shown in Fig. 4a – 4c, respectively. Hotspot temperature rise as coated coil at an applied bias of 20 V and it indicates the respective temperature points. Raman spectra and surface condition of GO on the coil are provided in Fig. 3c and 3d, respectively. As the carbon sp² lattice heavily defected during its synthesis process, GO appeared with a highly intense I(D) peak at ~1365 cm⁻¹, along with its broad I(G) peak at ~1591 cm⁻¹. The intensity ratio of I(D)/I(G) was 0.972. Noticeable stacks of GO sheets were observed from the optical microscopy image. Fig. 3e and 3f are the Raman spectra and surface condition of the G-GO-coated coil, respectively. After the Ar/CH₄ plasma treatment, defect peak intensity was increased with I(D)/I(G) of 1.277. In this case, G-GO is simply a highly reduced GO by the Ar/CH₄ plasma treatment. The G-GO film appeared darker than the original GO film.

Fig. 3a shows the photo of spin-coated GO on the coil. Similar to hBN, the thermograph in Fig. 3b shows the GO-
indicated by \( \frac{dT_0}{dP_{coil}} \) exhibited a significant cooling after the deposition of hBN and G-hBN. From the plots, we can obtain a coil cooling in terms of:

\[
\text{Coil cooling} = \frac{T_0 - T_1}{P_{coil}}
\]  

(1)

Fig. 4d shows the \( T_0 - T_1 \) versus \( P_{coil} \) for the hBN and G-hBN heat spreaders. For the device without a 2D heat spreader, the underlying SiO2 will act as a heat spreader for both lateral and vertical heat flow. The lateral heat flow will experience three \( \Theta \) across the coil, SiO2, and peripheral heat sinks, i.e. \( \Theta_{coil} \), \( \Theta_{SiO2} \), and \( \Theta_{sink} \) respectively. Considering the size of the peripheral heat sinks is sufficiently large, we suggest \( \Theta_{SiO2} + \Theta_{sink} \equiv \Theta_{SiO2} \). The \( \Theta \) for the lateral heat path can be written as:

\[
\Theta_{lateral} = \frac{T_0 - T_1}{P_{coil}}
\]  

(2)

The underlying SiO2 heat spreader will conduct the heat to the bottom and top peripheral heat sinks. Both heat sinks are mounted on the convective boundary to the ambient temperature. For the vertical heat path, the remaining \( \Theta_{Si} \) and \( \Theta_{stage} \) are not necessary for the discussion. After the deposition of the 2D heat spreader, the \( \Theta \) network will be modified accordingly. The difference between \( \Theta_{SiO2} \) and \( \Theta_{spreader} \) is in the heat spreading characteristic within the materials. For the isotropic \( \Theta_{SiO2} \), the temperature distribution should be uniform across the thickness at all vertical positions. While, for the anisotropic \( \Theta_{spreader} \), the heat travels laterally through the length of the heat spreader and then penetrates vertically through its thickness. More on this is described elsewhere [11]. The \( \Theta_{lateral} \) can be defined as \( \Theta_{SiO2}/\Theta_{spreader} \) based on a parallel circuit equation [12]. The desirable situation for hotspot cooling would be when \( \Theta_{SiO2}/\Theta_{spreader} \) and the value of \( T_0 \) should be reduced after the improvement in lateral heat spreading by the deposition of the 2D heat spreader.

Fig. 4e shows the \( T_0 - T_3 \) versus \( P_{coil} \) for the hBN and G-hBN heat spreaders. The reduction in hotspot temperature is tally with the improvement in coil cooling and reduction of \( \Theta \). The G-hBN is a superior heat spreader compared to hBN. Since the position of \( T_3 \) is above \( T_0 \), the value of \( d(T_0 - T_3)/dP_{coil} \) in Fig. 4f should be a re-verification for \( \Theta \). However, we found that \( d(T_0 - T_3)/dP_{coil} \) is prone to error since the emissivity of...
where

The Ar/CH\textsubscript{4} plasma treatment converts hBN and GO into materials changes from the bare coil, coil with hBN, and coil with G-hBN. While for the comparison, the \( T - P \) profile in terms of \( T_0 \) and \( T_1 \) are shown in Fig. 5a – 5c, respectively. We found a significant hotspot reduction \( \Delta T_0/dP_{\text{coil}} \) after conversion of GO into G-GO. Similar to the previous hBN and G-hBN heat spreaders, hotspot reduction is accompanied by an improvement in coil cooling in Fig. 5d and reduction in \( \theta \) in Fig. 5e. We avoid considering \( \theta \) by \( d(T_1-T_0)/dP_{\text{coil}} \) in Fig. 5f since the conversion of materials from GO into G-GO also requires material’s emissivity correction for the IR thermometer.

The comparison in both devices requires normalization to produce a fair comparison. The normalized parameter can be defined as:

\[
\Delta X = 1 + \frac{X_{\text{after}} - X_{\text{before}}}{X_{\text{before}}}
\]  

(3)

where \( X \) can be either coil resistance, \( R_{\text{coil}} \) (at \( T = 303 \text{ K} \), \( T_0 \), \( T_{\text{G}}-T_{\text{V}} \), or \( T_{\text{V}}-T_{\text{S}} \). Fig. 6a – 6d compares the changes in normalized parameters for hBN, G-hBN, GO, and G-GO heat spreaders. Even though the G-hBN and G-GO demonstrate the highest coil cooling of \( \Delta T_0 \), the highest coil cooling of \( \Delta T_{\text{G}} \) more than 0.5-times, the highest \( \theta \) of \( \Delta T_{\text{G}}-T_{\text{S}} \) more than 10-times, and the lowest \( \chi \) of \( \Delta T_{\text{G}}-T_{\text{S}} \) more than 0.5-times, the Ar/CH\textsubscript{4} plasma treatment significantly affected the \( \Delta R_{\text{coil}} \). The increment in \( \Delta R_{\text{coil}} \) after Ar/CH\textsubscript{4} plasma treatment to produce G-hBN and G-GO exceeds 1.2-times compared to the bare device, most probably due to the thinning of Pt/Cu/Ti coil by the physical plasma sputtering [13]. Using the same recipe for hybridizing hBN and GO into G-hBN and G-GO, the estimated etching rates by ions bombardment were at \(-2.07\text{ mm}^3\) for the Pt and SiO\textsubscript{2} substrate, respectively [13].

IV. CONCLUSION

The Ar/CH\textsubscript{4} plasma treatment converts hBN and GO into hybridized G-hBN and G-GO heat spreaders, respectively. Both G-hBN and G-GO were superior heat spreaders compared to the initial hBN and GO in terms of hotspot reduction, coil cooling, and reduction in lateral \( \theta \). However, the Ar/CH\textsubscript{4} plasma treatment induces damage to the device and the \( R_{\text{coil}} \) was severely increased. Therefore, the Ar/CH\textsubscript{4} plasma treatment may not be necessary. We found that the preferred direct contact 2D insulator heat spreader is hBN since it is the third-best after the G-GO and G-hBN, offering about 0.84-times \( \Delta T_0 \) (\(-16\% \) hotspot reduction), 0.63-times \( \Delta T_{\text{G}}-T_{\text{S}} \) (\(-37.4\% \) reduction in lateral \( \theta \)), and maintaining the initial value of \( R_{\text{coil}} \).