International Journal of Engineering, Management and Humanities (IJEMH)
Volume 6, Issue 1, Jan.-Feb, 2025 pp: 87-92 ISSN: 2584-2145 www.ijemh.com

Raman Analysis of Graphene Formation on Etched Si/SiO2 Substrates: Role of Nickel Layer Placement and Thermal Processing

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Date of Submission: 07-01-2025 Date of Acceptance: 19-01-2025

ABSTRACT: In this study, the direct formation of large-area graphene on Si/SiO_2 substrates composed of amorphous SiC or carbon films, positioned above or below a nickel (Ni) layer, using rapid thermal processing (RTP) at temperatures up to 1100°C. The primary objective was to investigate the effect of Ni layer thickness and positioning on the quality and coverage of graphene grown on SiC-based substrates. The RTP conditions exhibit that Ni over SiC samples give the highest quality graphene with approximately 40% surface area coverage, as confirmed by Raman spectroscopy. In contrast, Ni beneath SiC substrates produced graphene with 99% coverage, but the quality was lower. After etching the nickel silicide formed during the process, suspended graphene was obtained for thick Ni layers, while thinner Ni layers resulted in graphene retention on the Si/SiO_2 substrate. Raman spectra revealed the crucial role of a rapid cooling rate in promoting the formation of monolayer graphene.

KEYWORDS: *Graphene, rapid thermal processing, Ramn]an spectrum*

I. INTRODUCTION

Graphene has attracted significant attention for its remarkable electrical, thermal, mechanical, and optical properties. These exceptional characteristics make graphene an ideal material for a wide range of applications, including electronics, photonics, sensors, and composite materials [1-4]. However, despite its potential, the large-area and high-quality synthesis of graphene remains a major challenge, it must be grown on substrates that are compatible with existing semiconductor fabrication processes. The most widely explored methods for large-scale graphene synthesis include chemical vapor deposition (CVD) and epitaxial growth on silicon carbide (SiC) [1-4]. CVD-based methods have been successful in producing graphene on metal substrates such as nickel and copper [5-6]. While these methods can yield high-quality graphene, the process of transferring graphene from the metal substrates to device-compatible materials remains a significant barrier, limiting scalability for industrial applications. In contrast, epitaxial growth of graphene on SiC substrates has shown promise for fabricating electronic devices directly on the substrate [7-9]. However, this method often requires high temperatures, making it less suitable for certain applications, and it still faces challenges related to producing large-area graphene with uniform quality. A promising alternative is the use of rapid thermal processing (RTP) to synthesize graphene. RTP involves heating substrates to high temperatures in a short period, typically ranging from 750°C to 1100°C, to enable the growth of graphene from carbon sources such as nickel and silicon carbide. Recent studies have demonstrated that this method can produce high-quality graphene, often without the need for subsequent transfer steps [10,11]. This study aims to address these gaps by exploring the effect of the nickel layer's position—whether above or below the carbon or SiC layers—on the growth of graphene using RTP. We hypothesize that the positioning of the nickel layer, as well as its thickness, plays a crucial role in determining both the quality and coverage of the graphene produced. Additionally, we investigate how the cooling rate influences the formation of monolayer graphene. By employing Raman spectroscopy, we seek to correlate these factors with the structural properties of the resulting graphene, providing insights into the optimal conditions for large-area graphene synthesis without the need for transfer processes. Despite these advances, a critical gap in the literature exists regarding how the position and thickness of metal layers, such as nickel, affect the growth and quality of graphene during RTP. Several studies have focused on the effects of different growth temperatures, carbon sources, and substrates, but the precise role of the nickel layer configuration has not been fully elucidated [12-15]. Furthermore, while it

is well established that cooling rates influence the number of graphene layers formed, there is still a lack of comprehensive studies addressing how varying these rates impacts the formation of monolayer versus multilayer graphene. The aim of this study to direct growth of graphene large area on a Si substrate and SiO2 insulator layer. And to deposit a nickel layer in several samples using TPR.

II. METHODs and MATERIALs

A carbon film (SiC) and a metal film (Ni) sputtered onto a Si substrate. Deposition sequence: SiC/Ni or Ni/SiC. Then, rapid thermal processing (RTP) was conducted in the same chamber (slower heating, similar cooling) or in a separate machine (faster heating rates, reaching 1100°C instantly). Graphene consistently grows on top, regardless of deposition order. After RTP, dissolve Ni-silicide in HCl solution, letting graphene loosely adhere to the Si substrate. A SiO_2 layer is deposited on graphene, followed by bonding to a new insulator substrate. Remove the original substrate using a Si etcher and acid, ensuring good adhesion. Three samples were prepared as described previously (S1, S2, S3). The deposition process was carried out, and Raman spectra were tested before and after the etching process by HCl solution. The Raman spectra were compared and analyzed for the graphene layers.
The preparing condation were lested in Table 1.

Table 1: The deposion and RTP conadiations for three samples prepared

Samples Substrate: SiO_2	S1 C: 5nm Ni: 42nm	S2 C: 5nm Ni: 30nm	S3 C: 2.5nm Ni: 42nm
Befor etching RTP condition:	Intensity:(74%,1100°C); Heating & Cooling Rate: 15%; @: 3mins	Intensity:(74% at 1100°C); Heating & Cooling Rate: 15%; @: 3mins	Intensity: 74% (1100°C); Heating & Cooling Rate: 15%; @:3mins
After etching RTP condition	Intensity: 74% (1100°C); Heating and Cooling Rate: 15%; @: 3mins	Intensity: 74% (1100°C); Heating and Cooling Rate: 15%; @: 3mins	Intensity: 74% (1100°C); Heating and Cooling Rate: 15%; @: 3mins

The graphene growth process was illustration in the Fig. 1, the process shows that a carbon-containing (SiC) and (Ni) on a silicon wafer, in orders: SiC over Ni (a) or Ni over SiC (b). Next, an RPT step is performed, leading to the silicidation of Ni and the formation of graphene on top of the Ni silicides upon cooling (c). Following this, an insulating layer is sputtered directly onto the graphene, which is then bound to the silicon wafer (d). The sacrificial growth wafer is then removed (e). Alternatively, graphene can be transferred to the silicon wafer by dissolving the Ni silicide (f).

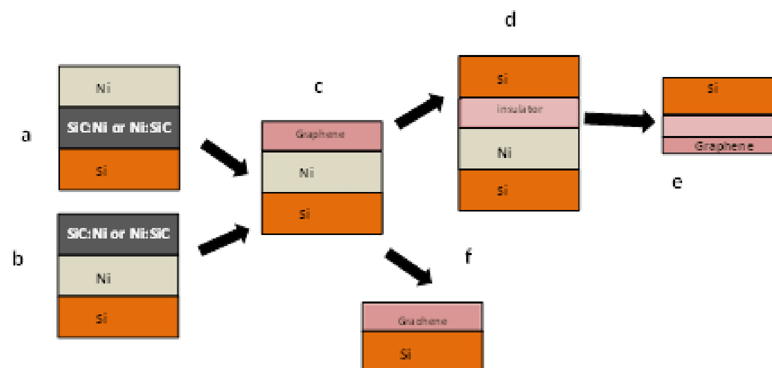
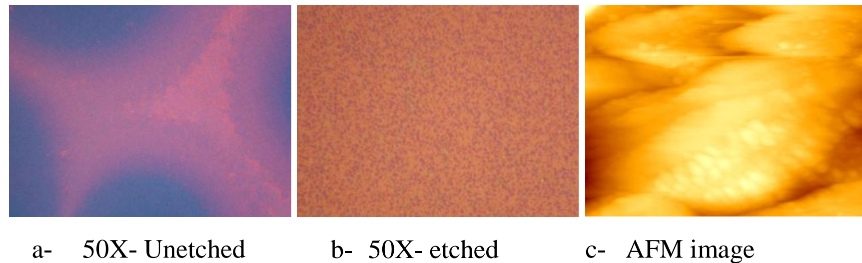


Figure 1: The sputter deposition process

III. RESULTS AND DISCUSSION

The results present a potential route for the production of large-area graphene directly on Si/SiO2 insulating wafers. Figures below show the growth mechanism and detailed characterisation of graphene by Raman, optical micrograph and AFM image of the sample.

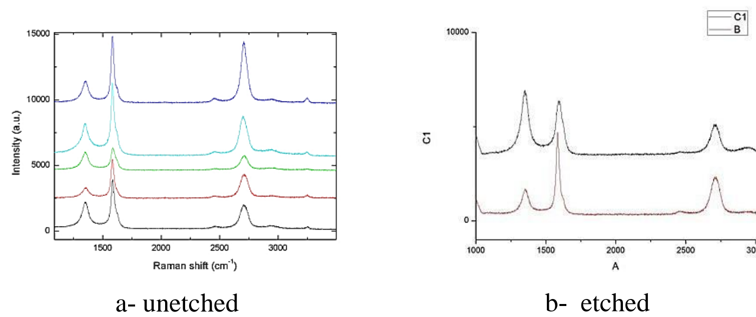


a- 50X- Unetched b- 50X- etched c- AFM image

Figure 2: The optical micrograph and AFM images of the S1 sample

Figure 2, for the sample S1 before etching, It shows the relative light area stands for more Ni islands or Ni particles on the surface, and the surface is very rough, we can see the grain size is about ~4nm, and it looks like a typical Ni grain size.

Figure 3, Raman spectrum for the sample S1 before etching (Fig. 3a) and after etching (Fig.3b).

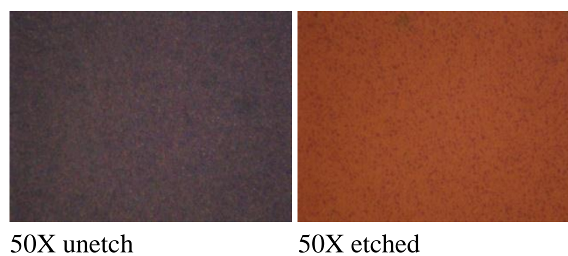


a- unetched b- etched

Figure 3: The Raman spectrum for S1 sample

The S1, 50X etched optical microscope revealed distinct color variations, indicative of varying graphene thicknesses. Regions devoid of the Ni film, appearing lighter, were observed. Despite the unknown location of Raman spectroscopy measurements, the light pink area, exhibiting a higher 2D/G ratio and lower contrast compared to the light area, is likely of higher graphene quality, as indicated by the red line. Conversely, the darker areas, characterized by similar shape, size, and density to the dark spots in the pre-etch image, displayed no significant improvement in quality after etching. Consequently, the location of the graphene layer relative to the Ni film (i.e., on top or underneath) remains inconclusive.

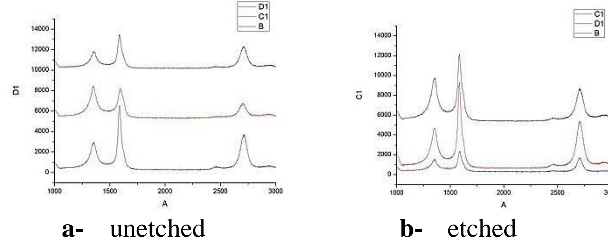
Figure 4, The optical microscope image for the sample S2 before etching (Fig.4a) and after (Fig.4b):



50X unetch 50X etched

Figure 4: The optical micrograph images of the S2 sample

Figure 5, The Raman spectrum for the sample S2 were shown, before etching in (Fig.5a) and after in (Fig.5b):

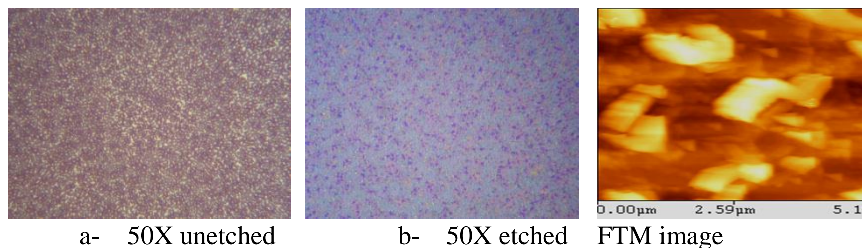


a- unetched **b-** etched

Figure 5: The Raman spectrum for S2 sample

In the S2 sample, prior to etching, numerous Ni islands were observed on the surface, readily visible under microscopy. This suggests incomplete evaporation of Ni or Ni carbide during the process. Notably, a significant increase in image brightness was observed after etching, consistent with the hypothesis that Ni or Ni carbide particles contribute to image contrast. Comparing S2 to S1, a higher D-band intensity was observed in S2, potentially attributed to a thinner Ni film. With a thinner Ni film, incomplete dissolution of carbon atoms into the Ni layer is likely, resulting in a higher concentration of amorphous carbon (a-C) on the surface and consequently, an elevated D-band intensity.

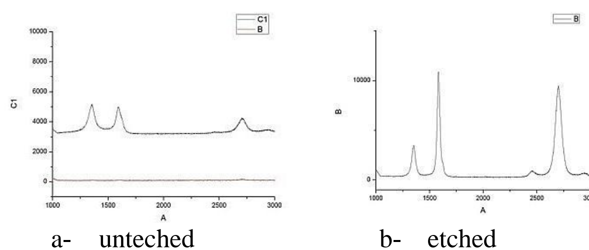
Figure 6, The optical microscope and AFM images for the sample S3 were shown, before etching in (Fig.6a) and after in (Fig.6b):



a- 50X unetched b- 50X etched FTM image

Figure 6: The optical micrograph images of the S3 sample and the AFM image

Figure 7, The Raman spectrum for the sample S3, before etching (Fig.7a) and after (Fig.7b):



a- unteched b- etched

Figure 7: The Raman spectrum for S3 sample

The 50X optical microscope image of the sample prior to etching exhibited a strong resemblance to that of S1, where all data was sourced from the original publication. The surface displayed a uniform green colouration, characteristic of sputtered graphene transferred onto a SiO_2 substrate, suggesting a similar Ni/C ratio to S1. In the 50X image before etching, a close resemblance to the published image was observed, with the notable exception of a significant residual Ni layer on the substrate, unlike the complete Ni evaporation reported in the literature, this is consistent with [16-18]. A 2D/G ratio of 1, indicative of bilayer graphene, was obtained, representing a significant improvement compared to the pre-etching result, strongly suggesting graphene growth beneath the Ni film. Although a decrease in the D-band intensity was observed, the presence of residual Ni islands, as evident in the S3 50X post-etch image, indicates potential for further reduction. Furthermore, a comparison with S1 revealed a superior graphene quality in S3,

corroborating the findings from S2, where an excess of carbon was attributed to an elevated D-band intensity. So the RTP conditions exhibit that Ni over SiC samples give the highest quality graphene with approximately 40% surface area coverage, as confirmed by Raman spectroscopy. In contrast, Ni beneath SiC substrates produced graphene with 99% coverage, but the quality was lower. This implies that a further reduction in the carbon content in subsequent samples may lead to a further decrease in the D-band intensity.

IV. CONCLUSION

This study demonstrates the feasibility of growing graphene on Ni silicides using a rapid thermal processing (RPT) approach. The results highlight the critical influence of Ni thickness and carbon content on graphene quality, with excessive carbon leading to increased D-band intensity, this is consistent with both [19,20]. Further optimization of the process, including improved Ni evaporation and precise control of carbon content, is necessary to achieve high-quality, large-area graphene for potential applications.

ACKNOWLEDGMENTS

Researchers here recall the opportunity provided by Wolfson Nanomagnetics Laboratory at Plymouth University-UK to work on the sidelines of day works to achieve this study. Also we thank the University of Mosul for giving us the opportunity to devote ourselves the sabbatical study.

REFERENCES

[1]. Geim, A. K., & Novoselov, K. S. (2007). The rise of graphene. *Nature Materials*, 6(3), 183-191.
[2]. Zhang, Y., Tan, Y. W., Stormer, H. L., & Kim, P. (2005). Experimental observation of the quantum Hall effect and Berry's phase in graphene. *Nature*, 438(7065), 201-204.
[3]. Li, X., et al. (2009). Chemical vapor deposition growth of graphene. *Journal of the American Chemical Society*, 131(43), 15939-15944.
[4]. Berger, C., et al. (2004). Electronic confinement and coherence in patterned epitaxial graphene. *Science*, 312(5777), 1191-1196.
[5]. Reina, A., et al. (2008). Large-area, few-layer graphene films on arbitrary substrates by chemical vapor deposition. *Nano Letters*, 9(1), 30-35.
[6]. Li, X., et al. (2011). Transfer of large-area graphene films for high-performance transparent conductive electrodes. *Nano Letters*, 10(10), 3951-3957.
[7]. Schedin, F., et al. (2007). Detection of individual gas molecules adsorbed on graphene. *Nature Materials*, 6(9), 652-655.
[8]. Kajiya, D., et al. (2009). Graphene growth on SiC by rapid thermal processing. *Journal of Crystal Growth*, 311(19), 4687-4690.
[9]. Eckmann, A., et al. (2012). A highly flexible graphene transistor. *Nano Letters*, 12(1), 373-378.
[10]. Huang, P. Y., et al. (2011). Grains and grain boundaries in single-layer graphene atomic patchwork quilts. *Nature*, 469(7330), 389-392.
[11]. Liao, L., et al. (2011). Epitaxial growth of large-area graphene on 4H-SiC(0001). *Carbon*, 49(12), 4141-4149.
[12]. Novoselov, K. S., et al. (2004). Electric Field Effect in Atomically Thin Carbon Films. Science, 306(5696), 666-669.
[13]. Bae, S., et al. (2010). Roll-to-Roll Production of 30-inch Graphene Films for Transparent Conductive Electrodes. Nature Nanotechnology, 5(8), 574-578.
[14]. Schwierz, F. (2010). Graphene Transistors. Nature Nanotechnology, 5(7), 487-496.
[15]. Hossain, M. R., et al. (2010). Growth of Epitaxial Graphene on Silicon Carbide: Current Status and Perspectives. Materials Science and Engineering: R: Reports, 71(4), 67-85.
[16]. Ohta, T., et al. (2007). Interlayer Interactions and Electronic Structure of Graphene Layers. Physical Review B, 76(8), 081404.
[17]. Starke, U., & Speck, J. S. (2017). Graphene on Silicon Carbide: Properties and Prospects. Materials Science and Engineering R: Reports, 118, 1-40.
[18]. Zhang, W., et al. (2014). Direct Growth of High-Quality Graphene Films by Rapid Thermal Processing. Applied Physics Letters, 105(13), 133106.

[19]. Wang, X., et al. (2010). Growth of Graphene from Solid Carbon Sources. Nature Materials, 9(11), 757-763.
[20]. Hensley, D.K., *et al*.(2015). Nickel-Catalyzed Growth of Graphene on a Silicon Carbide Substrate. Journal of Applied Physics,118(15), 155303.