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Research Article

Graphene Growth in Different Thickness by Chemical Vapor Deposition Method

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ABSTRACT

Graphene is a two-dimensional honeycomb material with an atomic-thick planar structure. Graphene is one of the most studied nanomaterials that can be used in nanotechnology applications. There are various methods for synthesizing or isolating graphene monolayers, but by far the most popular uses a process called chemical vapor deposition. Chemical vapor deposition, or CVD, is a process that has the potential to produce relatively high-quality graphene at scale. The CVD process is relatively straightforward with some specialized equipment. However, controlling gas volume, pressure, temperature, and timing is critical to producing good quality graphene. In this project, the synthesis of graphene was carried out at different temperatures, pressures and coating times to produce stable, controlled and durable graphene by chemical vapor deposition. The characteristics of graphene sheets obtained by SEM, AFM and Raman spectroscopy analyzes were determined, as well as the optimal parameters for a stable, sustainable and controlled production of graphene. In parallel, the electrical properties of graphene films on different thicknesses have been studied. Therefore, obtaining a thin film with suitable thickness, transmission and electrical properties of graphene, which is currently marketed worldwide, was investigated.

Keywords: *Graphene, Chemical Vapor Deposition, Nanotechnology, Materials science*

Kimyasal Buhar Biriktirme Yöntemi ile Farklı Kalınlıklarda Grafen Büyütülmesi

ÖZET

Grafen, atom kalınlığında düzlemsel altıgen bal peteği yapısına sahip iki boyutlu bir malzemedir. Nano teknolojik uygulamalarda kullanılabilme potansiyeli ile en çok çalışılan nano malzemelerden biri grafendir. Tek katmanlı grafen sentezlenmesinin veya izole edilmesinin farklı yolları vardır, ancak şu ana kadarki en popüler yöntem kimyasal buhar biriktirme adı verilen bir işlem kullanılmaktır. Kimyasal buhar biriktirme, CVD, yöntemi ile ölçekte nispeten daha yüksek kaliteli grafen üretme potansiyeline sahip bir metottur. CVD işlemi makul bir şekilde basittir, ancak bazı özel ekipman gerekli olmasına rağmen, iyi kalitede grafen oluşturmak için gaz hacimleri, basınç, sıcaklık ve süre parametrelerinin kontrolü büyük önem arz etmektedir. Bu proje çalışmasında Kimyasal Buhar Biriktirme Yöntemiyle stabil, kontrollü ve sürdürülebilir grafen elde edebilmek için farklı sıcaklık, basınç ve kaplama süreleri ile grafen sentezi gerçekleştirilmiştir. SEM, AFM ve Raman Spektroskopi analizleri ile elde edilen grafen tabakaların karakterizasyonu yapılmış ve kararlı, sürdürülebilir ve kontrollü grafen eldesi için optimum parametreler belirlenmiştir. Aynı zamanda grafenin farklı

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kalınlıklardaki filmlerinin elektriksel özellikleri incelenmiştir. Böylece tüm dünyada şu an ticarileştirilmeye çalışılan grafenin uygun alttaş üzerine büyütülmüş uygun kalınlıkta ince film eldesi, transferi ve elektriksel özelliklerinin incelenmesi yapılmıştır.

Anahtar Kelimeler: *Grafen, Kimyasal Buhar Biriktirme, Nanoteknoloji, Malzeme Bilimi*

I. INTRODUCTION

Nanomaterials, which have been created from nanoscale materials, are used in a wide variety of fields due to their exceptional properties [1, 2]. The most commonly used carbon nanomaterials are ideal candidates for advanced applications in electronics, membranes, wastewater treatment, batteries, capacitors, heterogeneous catalysis, and biological and medical sciences [2–7]. The most important reason why carbon-based nanomaterials are of great interest is their infinite bonding ability through various hybridizations in the arrangement of carbon atoms [8, 9]. Considered as the technology of the future, graphene is one of the most studied nanomaterials with its potential for use in nanotechnology applications. In 2004, Andrey Geim and Konstantin Novoselov obtained single-layer graphene for the first time using only adhesive tape (scotch tape method) [10–13]. Graphene, the first known two-dimensional material, is distinguished from other allotropes of carbon by its unique properties. The resulting graphene sheets were only a few atoms thick, but were stable and of high quality under ambient conditions. Geim called these layers monolayer crystalline films. They discovered that these films are two-dimensional semimetals with a very small distance between the conduction band and the valence band. They also discovered that these thin layers exhibit a strong ambipolar electric field effect. They showed that these films had electron and hole concentrations of up to 1013 per cm^2 [11]. Yu Zhang et al. experimentally studied the mechanism of magnetic conductivity of a micro-engineered monolayer of highly mobile graphene in 2005 and their experiments showed the quantum Hall effect, which is unusual for electrons and holes [14]. This extraordinary phenomenon of quantum conductivity in graphene is of interest for applications in carbon electronics and magnetoelectronics.

There are many different manufacturing methods for synthesizing graphene. The most widely used methods include micromechanical exfoliation, chemical vapor deposition (CVD), graphene oxide reduction, ultrasonic separation of graphene in selected solutions, and epitaxial growth [9]. The CVD process is a controlled growth process for the production of multilayer graphene on a large scale. Moreover, the CVD method is a widely used method for the production of carbon nanotubes and various nanomaterials [15–17]. The main mechanism of this method; saturation of the transition metal with carbon after exposure to hydrocarbon gas at high temperature [17]. These gas molecules effectively regenerate the carbon on the surface of the catalyst. In addition, catalysts play an auxiliary role for carbon atoms in the growth process. The solubility of the catalyst in carbon plays a very important role in controlling the number of graphene layers that grow by epitaxy. Using this method, a 30-inch rectangular single-layer graphene was fabricated [18]. Although this method is much more reliable and physically controllable for producing graphene on a large scale, it is a relatively expensive and complicated process. The main advantage of the process is its high compatibility with modern metal oxide and semiconductor technology [19].

The successful synthesis of a few layer graphene sheets by chemical vapor deposition was first reported in 2006 by P. Somani and M. Umeno [20, 21]. With this work, Somani opened a new door for a solution of some unsolved problems, such as controlling the number of layers and folding graphs. A nickel substrate used in this study, after chemical etching of a metal substrate, the graphene layers were separated and then transferred to another substrate. Somani et al. showed that graphene with several layers of thickness could be produced in this way without any complex chemical and mechanical processes. De Heer et al. from Georgia Institute of Technology successfully grew multiple layers of graphene epitaxially on single crystal silicon carbide [22]. They noticed that the charge carriers in the resulting layers differ from the charge carriers of graphite and exhibit the properties of

massless Dirac particles. They also discovered that epitaxial graphene has near-ballistic transfer properties. Unlike graphene sheets obtained by mechanical separation, the quantum Hall effect was not observed in epitaxial graphene. They stated that this effect was suppressed in bulk due to the lack of localized states of matter. It has been suggested that the resulting graphene could be suitable for nanoelectronics with low dispersion and high-speed operation at room temperature.

In 2013, Srikrishna et al. performed magnetoresistance measurements on multilayer graphene structures fabricated by CVD [23]. The magnetic properties were studied by measurements in two different geometries where the current is parallel to the surface and the current is perpendicular to the surface (CPP). Here, the large CPP magnetoresistance (MR) was measured due to the interaction between graphene and the metal substrate. This measured MR value cannot be explained by hitherto known MR mechanisms. Its simple manufacturing technology and ability to maintain an MR value even at room temperature make graphene structures ideal for magnetic field sensors, information storage applications, etc. At the same time, Srikrishna et al. indicate that the relationship between the stored item and the structure should be examined.

Zhang et al. from the University of California developed a graphene material on copper and nickel substrates using CVD in 2012 [24]. They investigated the importance of using graphene for flexible transparent conductors for organic photovoltaic (OPV) cells and field-effect transistors, and it was observed that graphene is better suited for both applications. They pointed out that despite significant advances in this research, there is still much to explore and understand. The number of publications and patents on graphene, which has been the subject of increased research since 2004, now exceeds one million. This huge interest in graphene research started immediately after the isolation of graphene in 2004 and has grown significantly over the years [25].

In this study, the factors affecting the quality and thickness of graphene, such as gas flow, gas ratio, gas flow time, reaction temperature, cooling rate, were studied by chemical vapor deposition process, and suitable parameters were found to achieve stable, controllable, durable, and high quality graphene. Thus, controlling and understanding graphene grown by CVD methods will greatly contribute to the literature, allowing it to be used more effectively in graphene-based nanoscience and nanotechnology applications.

II. MATERIALS AND METHODS

A. Materials

The growth of graphene was carried out at a chemical vapor deposition system; consists of a quartz tube 1 m long and 100 mm thick, a high temperature oven, 4 gas inlets and gas cylinders in a gas cabinet, two areas in which different temperatures can be reached and a vacuum system. High purity argon, hydrogen and methane gases connected to CVD system with stainless steel wires were used in this study. $CH_{4(g)}$ was used as a carbon precursor, $Ar_{(g)}$ was used as a carrier gas in the system due to its very stable structure and high thermal conductivity, and $H_{2(g)}$ acts as a reducing gas by removing metal oxide from the surface and also plays an important role in stabilizing the decomposition of hydrocarbon samples and changing the morphology of graphene. Copper foil used to produce graphene on a copper substrate had a thickness of 25 µm and a purity of 99.98% and was purchased from Sigma-Aldrich. Acetone and ethanol with EMSURE® quality were purchased from Merckmillipore which were used for preliminary step in preparation of Cu substrates. Si/SiO_2 dielectric substrates and PMMA were purchased from Sigma-Aldrich for transfer of thin films on Cu substrates.

B. CVD-growth Graphene

The copper sheet was cut into 1x1 cm^2 and 2x2 cm^2 pieces, after which many different pre-treatment tests were carried out to remove contaminants from the surface. Finally, according to the test results, all the films were cleaned with acetone and ethanol, then placed in an ultrasonic bath in deionized water. The use of ultrasonic baths at each stage of cleaning damages the films and does not allow obtaining a homogeneous coating. For this reason, an ultrasonic bath was used in the last step of the process. Substrates prepared to remove moisture from the samples were placed on quartz boats and placed in the reaction chamber of CVD system. After placing copper foil in the tube furnace, the high temperature furnace was set at 900°C, 950°C and 1000°C. Therefore, the surface morphology is expected to be changed by annealing the substrates at these temperatures, and thus graphene films with large surface areas could be achieved. Annealing parameters at different temperatures are given in Table 1.

Table 1 *Annealing parameters.*

Annealing Temperature *(^0C)*	**Annealing time** *(min)*	**H_2 flow rate** *(sccm)*	**Ar flow rate** *(sccm)*
900	20	10-50	200
	30	10-50	
950	20	10-50	200
	30	10-50	
1000	20	10-50	200
	30	10-50	

After the completion of the annealing process at given temperatures, 8 different sets (a total of 24 different experimental conditions) were formed by supplying methane gas at different pressures, different CH_4 flow rates and different times to obtain graphene films (Table 2). Afterwards, the cooling process was completed and graphene films were obtained on the targeted substrates.

Table 2. *Coating parameters.*

No	**Coating Pressure** *(mbar)*	**Coating Temperature**	**CH_4 Flow rate** *(sccm)*	**H_2 Flow rate** *(sccm)*	**Ar Flow rate** *(sccm)*	**CH_4 Coating time** *(min)*	**Annealing time** *(min)*
1a	5.10^2	900^0C	5	50	200	5	20
1b	5.10^2	900^0C	5	50	200	10	20
1c	5.10^2	900^0C	5	50	200	15	20
2a	**5.10^2**	**900^0C**	**10**	**50**	**200**	**5**	**20**
2b	**5.10^2**	**900^0C**	**10**	**50**	**200**	**10**	**20**
2c	**5.10^2**	**900^0C**	**10**	**50**	**200**	**15**	**20**
3a	5.10^2	900^0C	15	50	200	5	20
3b	5.10^2	900^0C	15	50	200	10	20
3c	5.10^2	900^0C	15	50	200	15	20
4a	**5.10^2**	**950^0C**	**5**	**50**	**200**	**5**	**20**
4b	**5.10^2**	**950^0C**	**5**	**50**	**200**	**10**	**20**
4c	**5.10^2**	**950^0C**	**5**	**50**	**200**	**15**	**20**
5a	5.10^2	950^0C	10	50	200	5	20
5b	5.10^2	950^0C	10	50	200	10	20
5c	5.10^2	950^0C	10	50	200	15	20
6a	**5.10^2**	**950^0C**	**15**	**50**	**200**	**5**	**20**
6b	**5.10^2**	**950^0C**	**15**	**50**	**200**	**10**	**20**
6c	**5.10^2**	**950^0C**	**15**	**50**	**200**	**15**	**20**
7a	6.10^2	900^0C	25	10	200	10	30
7b	7.10^2	900^0C	25	10	200	20	30
7c	5.10^2	900^0C	25	10	200	30	30
8a	**6.10^2**	**1000^0C**	**5**	**50**	**200**	**15**	**20**

8b	**6.10^2**	**1000^0C**	**10**	**50**	**200**	**15**	**20**
8c	**6.10^2**	**1000^0C**	**15**	**50**	**200**	**15**	**20**

Graphene coated on copper films were transferred to Si/SiO_2 dielectric substrates. In this transfer, graphene films were first coated with PMMA using a centrifugation process. Indeed, graphene films are not damaged by acid etching. PMMA-coated samples were dried on a hot plate and then etched with acid suitable for the substrate type (etching) to separate the metal substrates from the graphene film. The separated graphene films were washed with distilled water and HCl and moved onto Si/SiO_2 substrates. As the final step, PMMA was removed from the graphene film surface.

Characterizations of graphene films were investigated through a number of analyses. The shape, smoothness and number of layers of graphene films were measured using an optical microscope, Raman spectroscopy, atomic force microscope (AFM), scanning electron microscope (SEM) and their electrical properties were measured using a dielectric measurements.

III. RESULTS AND DISCUSSION

A. Scanning Electron Microscopy (SEM) Analyses

CVD-graphene on copper substrates coated at 900^0C with CH_4 flow rate of 5 sccm, 10 sccm, 15 sccm, and coating time 5 min, 10 min and 15 min (Sample No:1a,2a,3a, see Table 2) SEM images under 5 μm, and 200 μm magnification is given in Figure 1 and Figure 2, respectively.

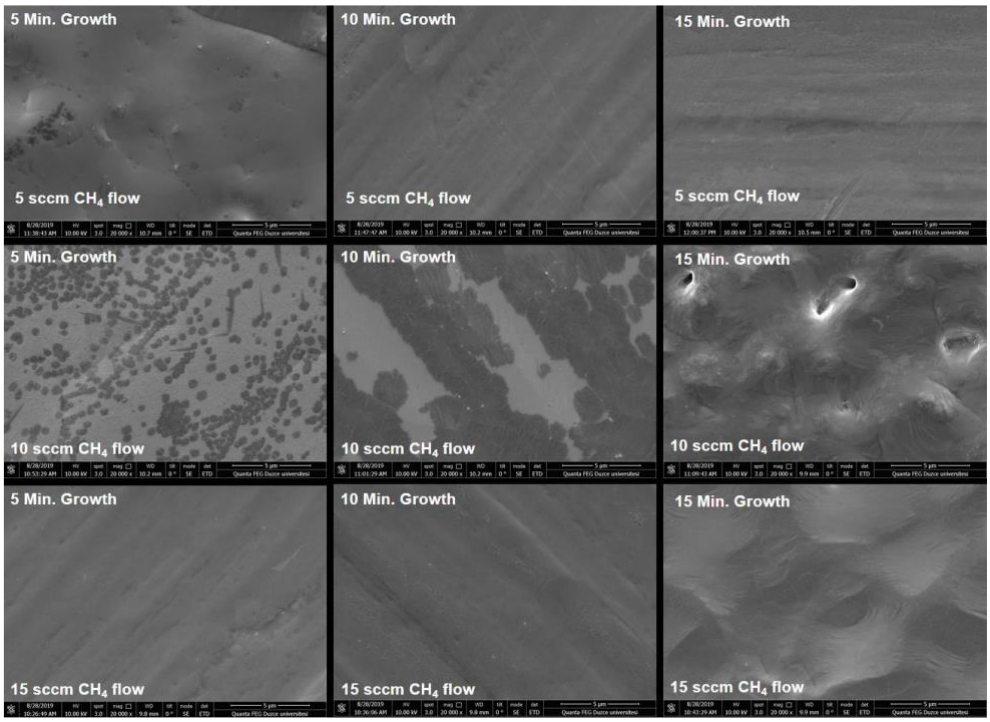

Figure 1. *CVD-graphene on copper substrates coated at 900^0C with CH_4 flow rate of 5 sccm, 10 sccm, 15 sccm, and coating time 5 min, 10 min and 15 min SEM images under 5 μm magnification.*

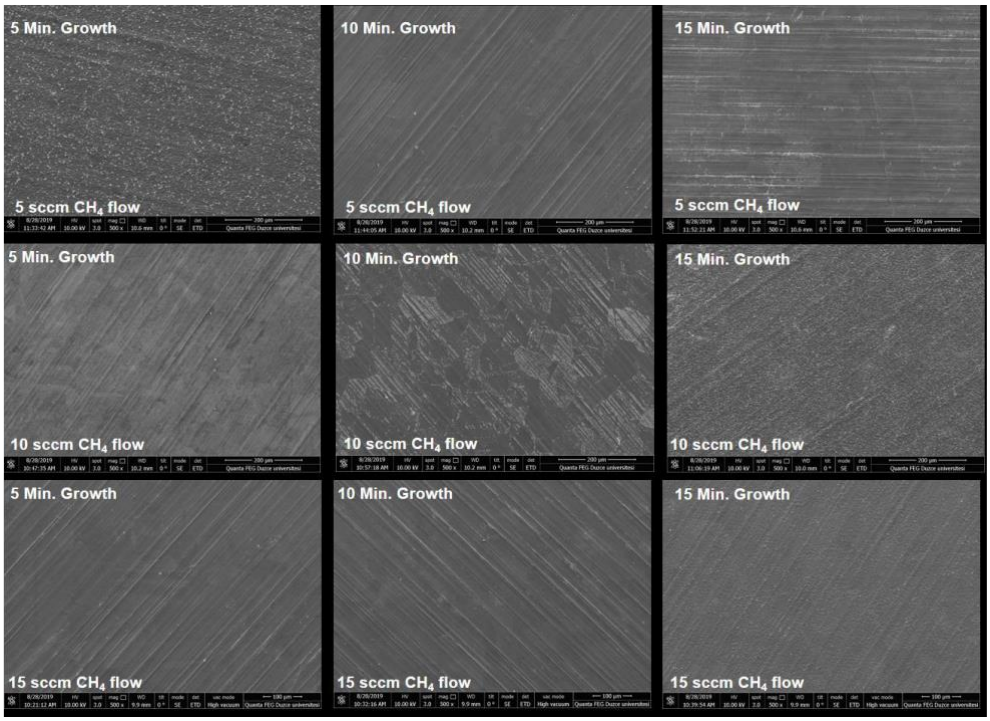

Figure 2. *CVD-graphene on copper substrates coated at 900^{0}C with CH_4 flow rate of 5 sccm, 10 sccm, 15 sccm, and coating time 5 min, 10 min and 15 min SEM images under 200 μm magnification.*

CVD-graphene on copper substrates coated at 950^{0}C with CH_4 flow rate of 5 sccm, 10 sccm, 15 sccm, and coating time 10 min, and 15 min SEM images under 5 μm, and 200 μm magnification is given in Figure 3 and Figure 4, respectively.

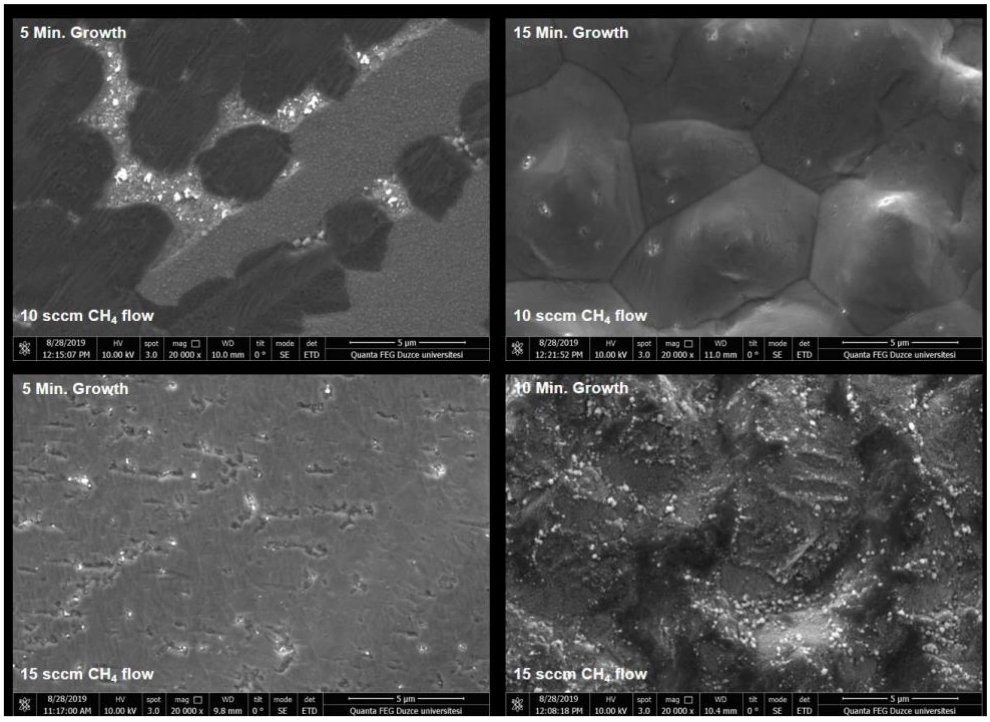

Figure 3. *CVD-graphene on copper substrates coated at 950^{0}C with CH_4 flow rate of 5 sccm, 10 sccm, 15 sccm, and coating time, 10 min and 15 min SEM images under 5 μm magnification.*

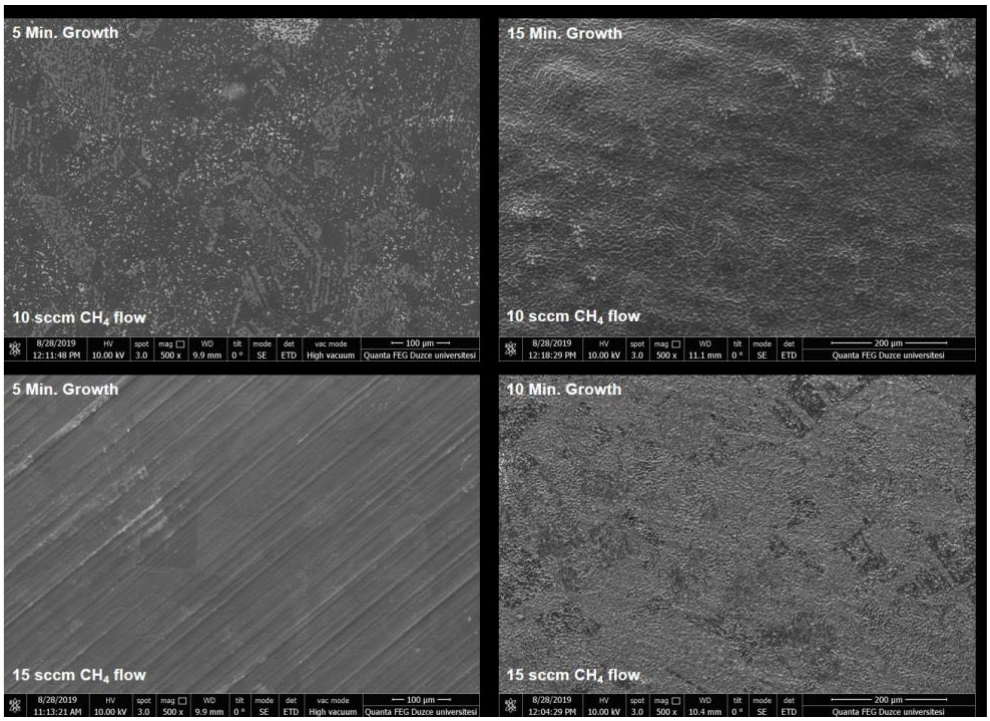

Figure 4. *CVD-graphene on copper substrates coated at 950^{0}C with CH_4 flow rate of 5 sccm, 10 sccm, 15 sccm, and coating time, 10 min and 15 min SEM images under 200 µm magnification.*

The samples given in Table 2, all of the experiments numbered 1(a, b, c), 2(a, b, c), 3(a, b, c) of the sets were successful. Experiments 4(b, c), 5(c) and 6(b) were successful. While there were local burns on the surfaces of the samples produced with 4(a), 5(a, b) coating parameters, all of the 6(a, c) samples were burned. All of the samples in group 8 were burned and the coating did not take place. In Figure 2, SEM image of 5 sccm CH_4 flow and 5 min coating time, it is seen that there are variety of impurities on coating surface. These impurities would be calcium silicate ($CaSiO_3$). This compound is known to be formed by silicon atoms falling from the quartz reactor when the remaining calcium atoms in the copper foils rise to the surface at high temperature. These impurities prevent the continuation of graphene, make it difficult to transfer and degrade its properties[26].

B. Atomic Force Microscope (AFM) Analyses

AFM analysis of CVD graphene samples was performed for the topological examination of the films. In Figure 5a, AFM analysis of monolayer graphene on Cu film is given as a reference. In Figure 5b, AFM analysis of the synthesized CVD graphene sample is given for comparison with the reference image. The results showed that the CVD-graphene samples were compatible. The carrier charge density of the single-layer CVD-Graphene film is 4000 $cm^2V^{-1}s^{-1}$ (reference sample from Sigma-Aldrich). The carrier charge densities of the CVD graphene films we prepared are in the range of 1000-4000 $cm^2V^{-1}s^1$. Prepared samples (Table 2) of CVD-graphene on copper substrates Sample 1b is single layer however graphene film growth is not uniform entirely, Sample 2b is single layer, Sample 6c is multi layer (up to 10layers). As the thickness of the graphene decreases, the carrier charge density increases. As a result, increasing the CH_4 flow rate and coating time decreases the carrier charge density.

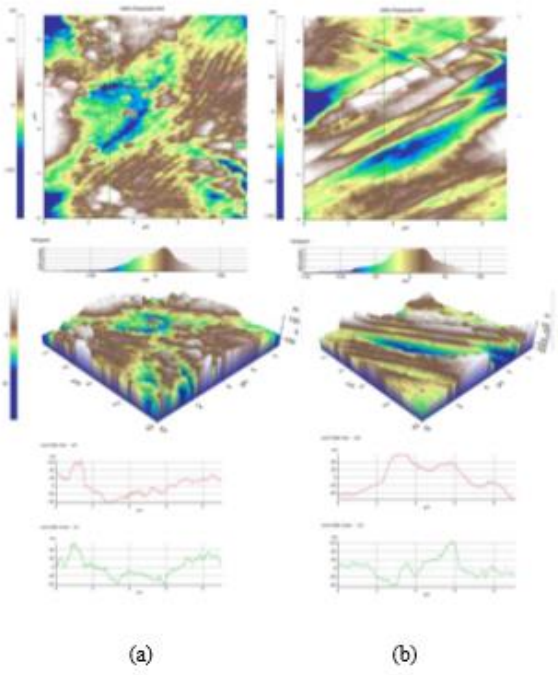

Figure 5. *AFM anayses of CVD-graphene sample*

C. Optical Microscope and Raman Spectroscopy Analyses

The optical microscopy image of CVD graphene films at 900°C with a flow rate of 10 sccm and a coating time of 10 minutes at 100x magnification is shown in Figure 6. Areas coated with monolayer graphene appear light orange and shift to blue as the number of layers increases.

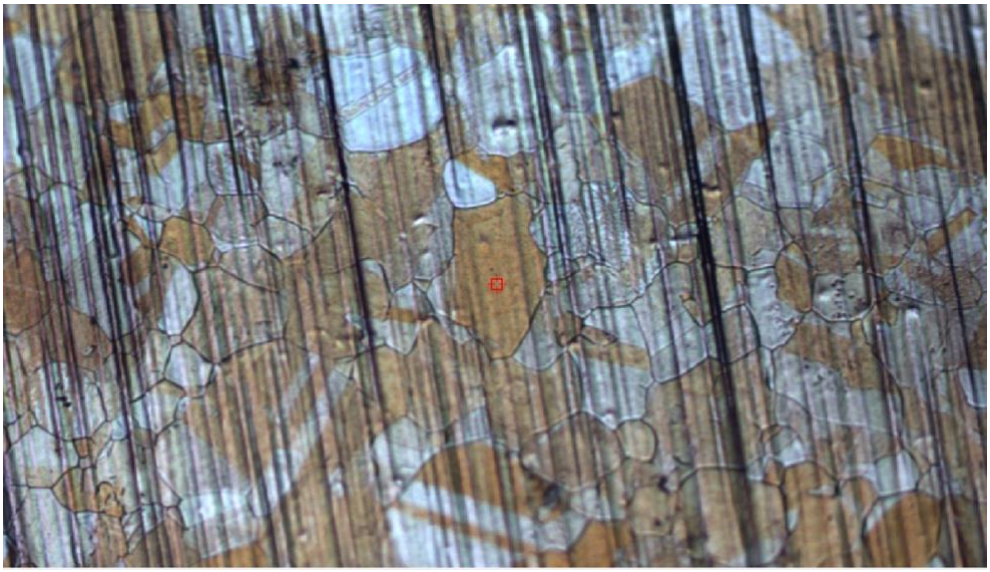

Figure 6. *The optical microscopy image of CVD graphene films at 900°C with a flow rate of 10 sccm and a coating time of 10 minutes at 100x magnification*

All samples were visualized with an optical microscope and also examined with a Raman spectrometer. The surface image was examined with an optical microscope and a color change was

observed as the structure changed from a single layer to a multilayer. The orange areas are single-layered and the blue ones are multi-layered (up to ten layers).

The D-, G- and 2D peaks are important for us in Raman spectrometry. Of these peaks, 2D and G indicate sp^2 hybridization at carbon atoms, and the absence of a D peak indicates a highly crystalline surface. The I_{2D}/I_G ratio provides information on the thickness of the graphene films and the number of layers. If the I_{2D}/I_G ratio is less than 1, this indicates the formation of a multilayer film. Figure 7 shows that the graphene film is approximately two to three layers thick. The visible D band in the graph shows that there are defects in the structure.

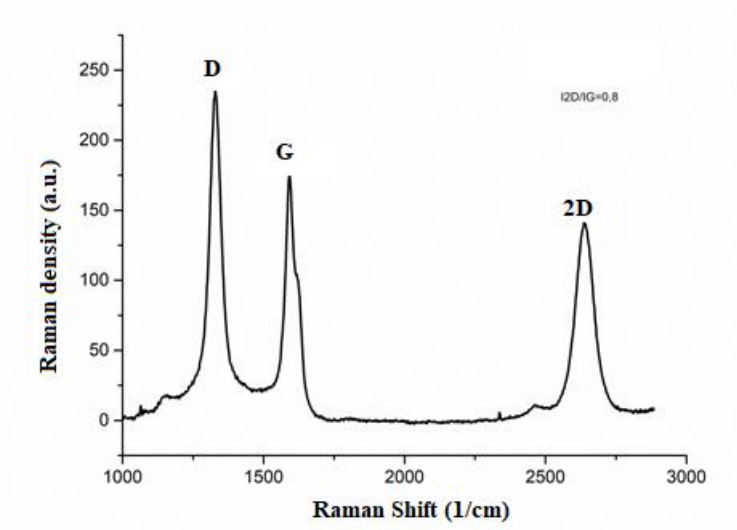

Figure 7*. Raman Spectrum of CVD graphene films at 900°C with a flow rate of 10 sccm and a coating time of 10 minutes*

IV. CONCLUSION

The chemical vapor deposition (CVD) method is a controllable growth method to produce large-area several-layer graphene. Although this method is much more reliable and physically controllable for producing graphene over large areas, it is a relatively expensive and difficult method. The biggest advantage of the method is its high compatibility with today's modern metal oxide semiconductor technology. In the formation of graphene films on copper substrate by CVD method diffusion mechanism will takes place. It is thought that the dissociated carbon atoms stay on the metal surface and become graphene directly. Although there have been many studies done so far, graphene formation mechanisms are still an issue that needs to be studied and studied.

Optical imaging results from graphene films are compatible with the literature[27,28]. It was observed that single-layer graphene films were orange, while a blue shift was observed as the number of layers increased. SEM results revealed that the number of impurities is very high on the sample where the CH_4 flow rate is 5 sccm and the coating time is 5 minutes. CVD-graphene films produced under 15 sccm methane flow rate obtained CVD-graphene films more than 10 layers, thus resulting in three-dimensional graphite structure. 1000ºC coating temperature is not suitable for CVD-graphene production because at 1000ºC to obtain CVD-graphene methane flow rate should be higher than 15 sccm. Therefore, 900ºC coating temperature is more convenient to obtain single layer and multilayer (up to 10) CVD-graphene thin films. It has been reported that these impurities are calcium silicate. It is known that when the calcium atoms remaining in the copper foils come to the surface at high temperature, this compound is formed with the silicon atoms raining from the quartz reactor. These impurities prevent the continuity of graphene, make its transfer difficult and affect its properties.

Despite the contamination on the CVD-Graphene films coated at 900^0C, the coating took place. However, it has been observed that some films prepared at 950^0C burned. The AFM results from the samples were compared with the monolayer graphene results, as the reference sample. Our monolayer samples are in agreement with the reference. The carrier charge density of the single-layer CVD-Graphene film is 4000 $cm^2V^{-1}s^{-1}$ (reference sample from Sigma-Aldrich). The carrier charge densities of the CVD graphene films we prepared are in the range of 1000-4000 $cm^2V^{-1}s^{-1}$. As the thickness of the graphene decreases, the carrier charge density increases. As a result, increasing the CH_4 flow rate and coating time decreases the carrier charge density. Raman Spectroscopy is an important tool for characterization of graphene due to the D, G and 2D peaks. 2D and G peaks indicate sp^2 hybridization at carbon atoms, while the absence of the D peak indicates a highly crystalline surface. The I_{2D}/I_G ratio gives information about the thickness of the graphene films and the number of layers. If the I_{2D}/I_G ratio is less than 1, it indicates that a multi-layered film is formed. Figure 7 shows that the graphene film is approximately two to three layers thick. The visible D band in the graph shows that there are defects in the structure. When the Raman spectrometers of the prepared graphene films were examined, it was seen that the number of layers changed depending on the methane flow rate. A bilayer structure was obtained in the coatings with a methane flow rate of 5 sccm. It has been observed that as the methane flow rate and time are increased, the structures turn into multi-layered structures and graphite coating is obtained in some films. While making the Raman analysis of the prepared films, both the front surface and the back surface were examined. According to the results obtained, graphene film was formed on both surfaces. It was observed that a two-layered structure was formed on the front surface (upper surface) and a multi-layered structure was formed on the back surface (<10 layers) when methane flow rate of 5 sccm. 5 sccm methane flow rate is sufficient for 1x1 cm^2 copper substrates, while that flow rate insufficient for formation of graphene on 2x2 cm^2 Cu-substrate. Higher methane flow rate (15 sccm) is enough to obtain CVD graphene. Thus, High CH_4 flow rate is more convenient to obtain large area CVD-graphene.

In conclusion a number of graphene films were obtained with the CVD method with different experimental and preliminary conditions, and measurements were taken from 24 of them. The prepared graphene films were then transferred onto Si/SiO_2 dielectric substrates. During this transfer, it was observed that the quality of the graphene films deteriorated and there were breaks. It is planned to focus on the following principle in the studies to be carried out in the future. To produce more homogeneous thin films and secondly to grow directly on the dielectric substrate without the need for any intermediate processing to prevent transfer losses. The measurement results confirmed that we obtained single-layer, two-layer and multi-layer graphene films. Our studies will continue in the form of obtaining graphene for application areas.

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