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## Improvement of Photo-Catalytic Properties of Graphene by N-Doping

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**Abstract:** Graphene is a two-dimensional, honeycomb-structured material consisting of a uniform monolayer of carbon atoms. Most scientists believe that graphene has revolutionary properties and research on graphene is increasing exponentially. It is considered as an alternative material that can replace silicon according to its properties, and graphene is called the supermaterial of the next century. On the other hand, some applications of graphene require the rearrangement of electronic properties. The chemical doping method is one of the most frequently used methods for this arrangement. In this study, nitrogen-doped graphene was synthesized by chemical vapor deposition (CVD) method. Urea was used as a nitrogen-doping source. Different amounts of urea were used to optimize the chemical nitrogen doping process. Raman spectroscopy was used for the characterization of the synthesized nitrogen-doped graphene. During the experiments, light (400-700 nm wavelength) was sent to the nitrogen-doped graphene surface with a solar simulator (A-type 150W, 1-3 SUN, Xenon lamp, 1.5 AM Filter). The photo-catalytic properties of the samples were measured by the potentiostatic technique.

**Keywords:** Energy, Graphene, Lithium, Battery

### Introduction

Since its first fabrication by mechanical exfoliation in 2004, graphene, a single-atom-thick sheet of carbon, has drawn significant interest from the scientific and technical community (Novoselov et al., 2004). Graphene's distinctive 2D honeycomb lattice structure gives it several exceptional chemical and physical capabilities in addition to making it the thinnest and strongest material in the universe (Geim & Novoselov, 2007).

Heteroatom doping can be used to successfully alter the structures and characteristics of carbonaceous materials. In accordance with this theory, doping graphene with nitrogen (N) can drastically alter the characteristics and functionality of graphene-based materials. By adding more heteroatoms, it is possible to alter the band gap of graphene, causing it to exhibit semiconducting qualities. The catalytic activity of doped graphene toward electro-catalytic or photo-catalytic processes can be considerably increased by the development of active areas by asymmetric spin and charge distributions during heteroatom doping in the graphene network (Wang et al., 2012).

In the study, it was aimed to optimize the photo-catalyst properties of nitrogen-doped graphene synthesized by chemical vapor deposition (CVD) method and then to use n-doped graphene as a photo-electrode in a Li-ion oxygen battery.

## Method

N-doped graphene was produced by evaporation of urea ( $\text{CH}_4\text{N}_2\text{O}$ ) powder in the chemical vapor deposition method. Methane ( $\text{CH}_4$ ) was used as a carbon source in the formation of graphene. The production flow chart of n-doped graphene is given in Figure 1.

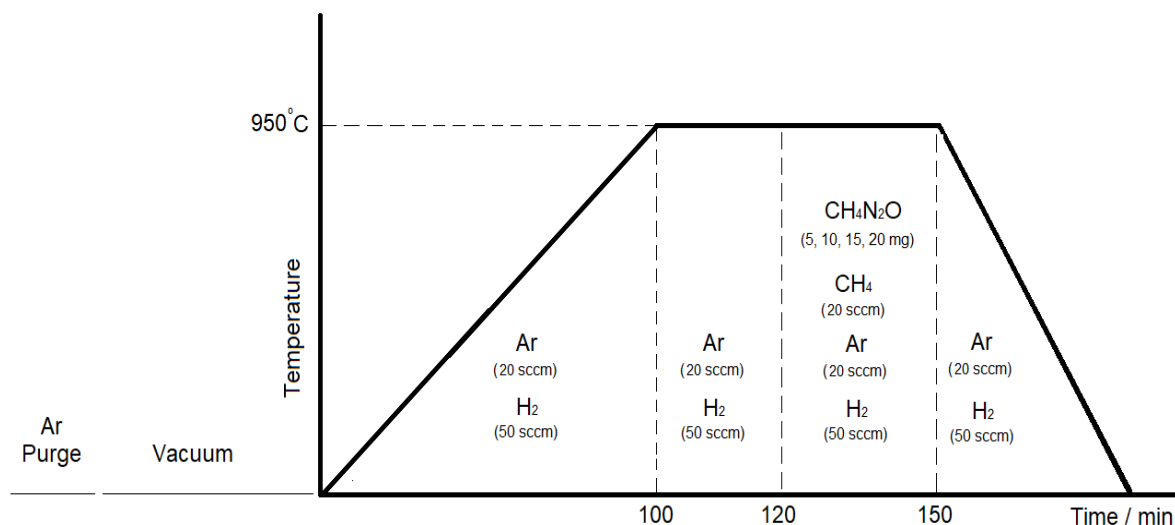


Figure 1. Production flow chart of n-doped graphene

Urea powder was evaporated by drawing it from the cold region of the quartz cell in the chemical vapor deposition furnace during the graphene growth stage to the region where the temperature varies between 60°C and 70°C, thus providing a highly controlled production. In studies conducted for this purpose, the most appropriate urea amounts were determined as 5 mg (U5), 10 mg (U10), 15 mg (U15) and 20 mg (U20) per  $\text{cm}^2$ , and comparisons were made on these 4 different compositions. Graphene is produced on copper substrates. Afterward, copper was dissolved in 50% HCl solution containing 0.5 M  $\text{FeCl}_3$ . Finally, graphene was cleaned with acetone/ethanol combinations.

During the experiments, light (400-700 nm wavelength) was sent to the nitrogen-doped graphene surface with a solar simulator (A-type 150W, 1-3 SUN, Xenon lamp, 1.5 AM Filter). The photo-catalytic properties of the samples were measured by the potentiostatic technique. Samples were examined with RENISHAW RAMAN inVia Microscope brand Raman Spectroscopy. Raman measurements were made using a laser at a wavelength of 532 nm.

## Results and Discussion

### Raman Spectroscopy

A highly helpful technique for describing N-doped graphene is Raman spectroscopy. The main characteristics of the spectrum of N- graphene are the D, G, and 2D bands. The two main peaks of graphene, 2D ( $\sim 2700 \text{ cm}^{-1}$ ) and G ( $\sim 1580 \text{ cm}^{-1}$ ), show  $\text{sp}^2$  hybridization at carbon atoms (Ferrari et al., 2006). The D peak ( $\sim 1350 \text{ cm}^{-1}$ ) observed in addition to these two peaks indicates the presence of  $\text{sp}^3$  carbons in the structure, in other words, the defects in the structure (Ferrari et al., 2006). Observation of the 2D peak without the D peak indicates a highly crystalline surface. Small shifts in peak positions are due to differences in synthesis temperature. The ratio of the 2D peak to the G peak intensity gives information about the number of graphene layers. If this ratio is above 2, it is accepted that the graphene is in a single layer, and the number of layers increases as the ratio decreases.

Raman spectra of nitrogen-doped graphene produced with 5 mg, 10 mg, 15 mg and 20 mg urea powder are presented in Figure 2. As a result of the analysis, D ( $\sim 1350 \text{ cm}^{-1}$ ) peak was also detected. The presence of this peak, which belongs to the defects in the structure, indicates that nitrogen doping has taken place. It was also observed that as the amount of urea increased, the relative ratio of the D peak to the 2D and G peaks also increased.

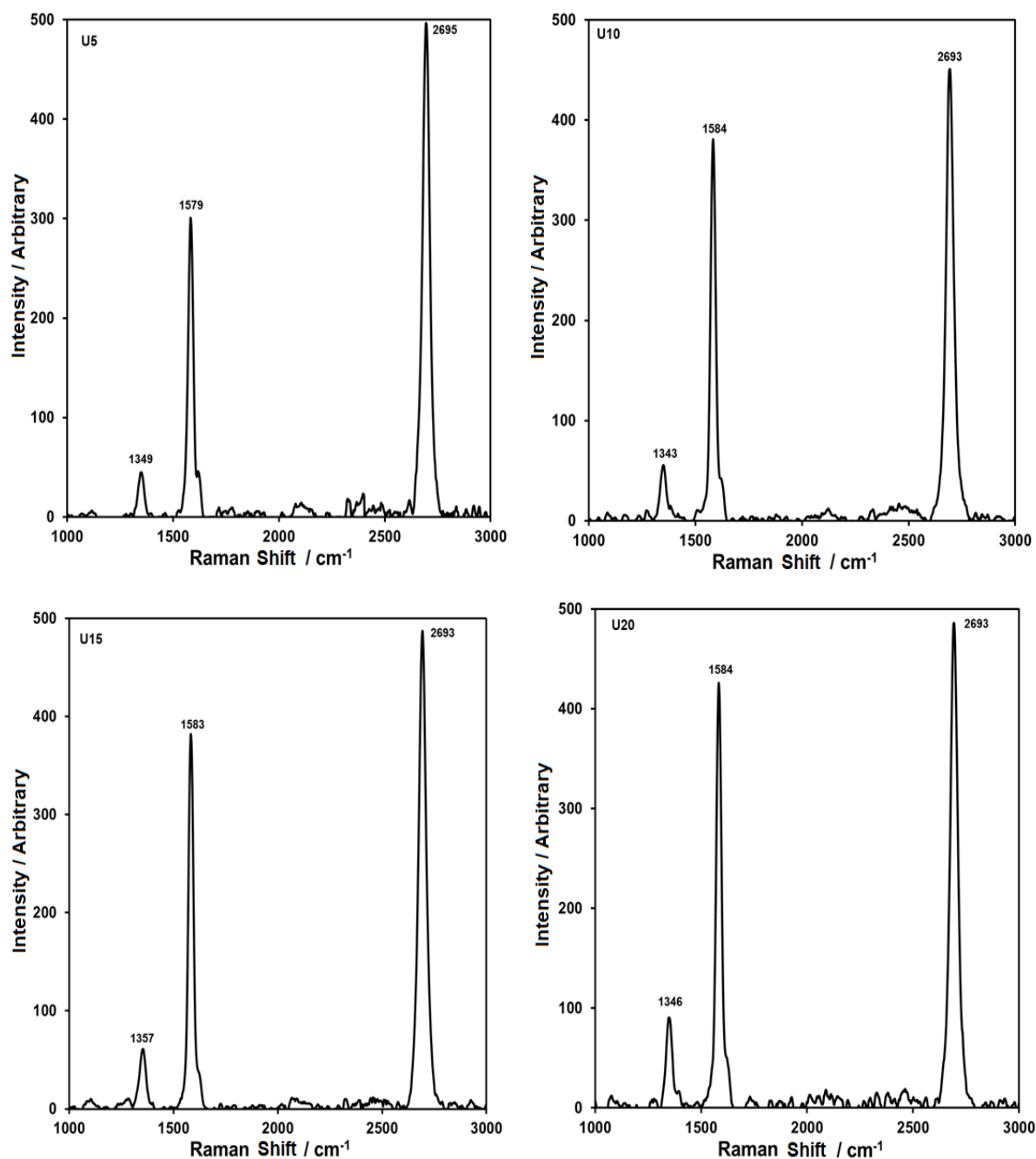


Figure 2. Raman spectra of n-doped graphene produced with 5 mg, 10 mg, 15 mg and 20 mg urea powder

### Optical Properties

The optimum amount of urea for nitrogen-filled graphene to be used as photo-electrode was tried to be determined by using photo-currents. The optimum amount of urea was determined by reference to such data for both characterization and performance experiments. The photo-anodic current measurements were obtained at a very low value of 50 mV anodic overpotential. Despite this, the obtained photo-anodic current values are at a remarkable level. This exceptional-electrode performance has been achieved by depositing the semiconductor homogeneously on the electrode surface.

When evaluated depending on the amount of urea powder used in the nitrogen-filled graphene synthesis process, the highest photo-anodic currents were obtained with 10 mg of urea powder (U10). Photo-current values of the U10 sample are presented in Figure 3.

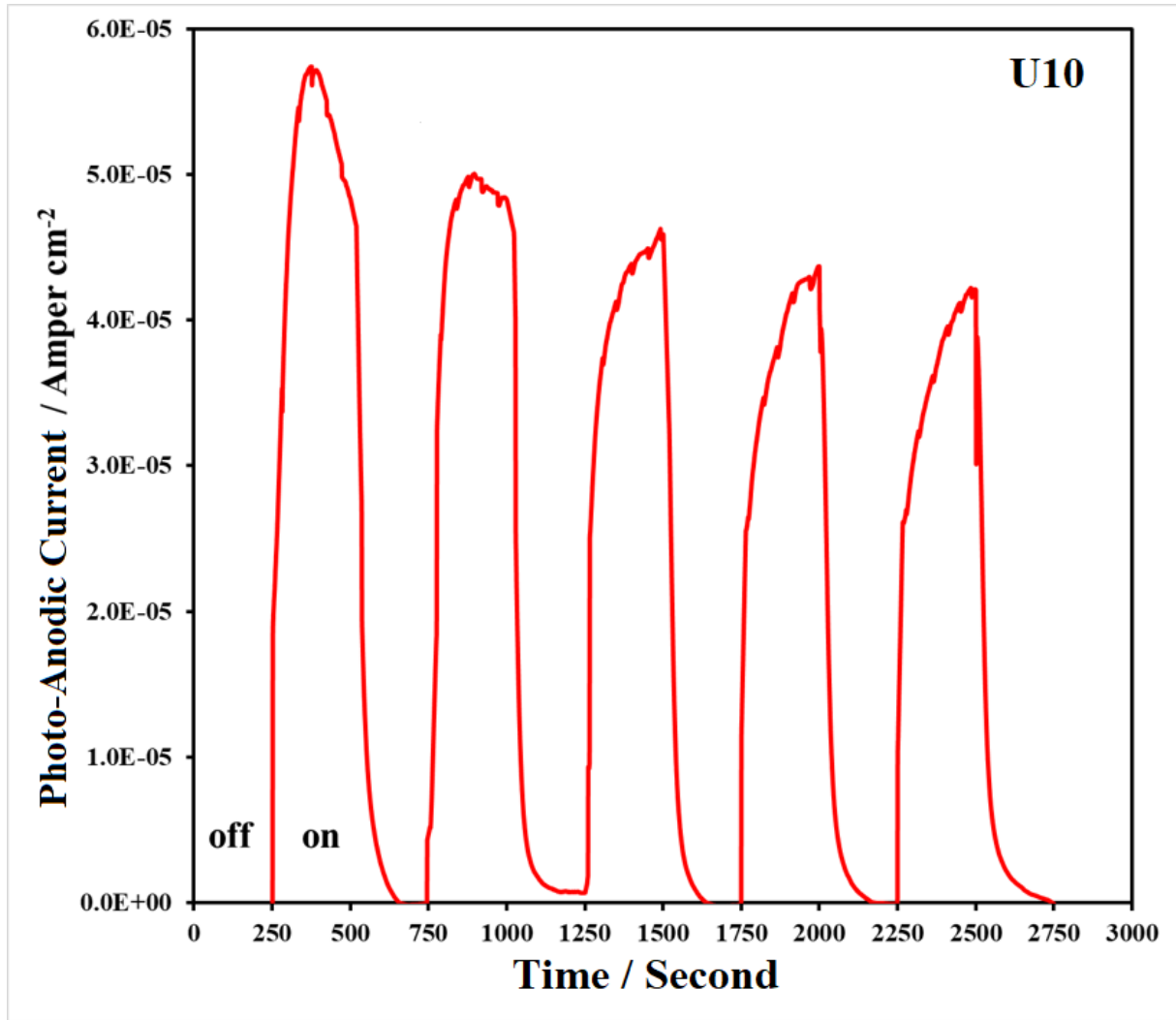


Figure 3. Photo-current values of the U10 sample are presented

## Conclusion

In the study, it was determined that the n-doped graphene obtained by evaporation of 10 mg of urea powder during the graphene growth process per 1 cm<sup>2</sup> copper substrate produced the highest photo-current. It has been observed that nitrogen doping contributes more to the photo-catalytic properties in the pyridinic, pyrrolic form compared to the graphitic form, provided that the structural integrity of graphene is preserved.

## Scientific Ethics Declaration

The authors declare that the scientific ethical and legal responsibility of this article published in EPSTEM journal belongs to the authors.

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