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Synthesis of Nanocomposite Photo-Catalysts for Photo-Assisted Charging of Li-Ion Oxygen Batteries

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Abstract: In this work, g-C_3N_4 / 3 % rGO nanocomposite was synthesized as a photo-catalyst in order to use in the Li-ion oxygen battery. The aim was to reduce the high charging potential of the battery by the photo-assistance. The synthesis of nanocomposite was carried out by thermal decomposition of melamine which was initially mixed with the required amount reduced graphene oxide (rGO). The rGO had dual competing actions in the nanocomposite. The first action was to reduce the optical band gap of the semiconductor nanocomposite that the photo-catalyst properties of the nanocomposite were improved. Secondly, rGO degradated the visible light utilization of the nanocomposite since it favorably absorbed incident light instead of g-C_3N_4. The photo-assisted charging tests indicated that the synthesized nanocomposite reduced the charging potential and improved the cyclic discharge-charge performance of the Li-ion oxygen battery.

Keywords: Nanocomposites, Photo-charging, Li-Ion oxygen batteries

Introduction

Li-ion oxygen batteries have one important drawback in the long way to the commercialization that they have unacceptably high charging potential (4.5 $V_{Li+/Li}$) due to the sluggish oxidation kinetics of the low conducting discharge product (Li_2O_2) (Lu, 2011). It is proposed that a photo-assisted charging of the Li-ion oxygen batteries by integrating a photo-electrode with the aid of triiodide/iodide (I_3^-/I^-) redox shuttling reduce the charging potential down to the discharging potential levels (2.8 $V_{Li+/Li}$) (Yu, 2014).

g-C_3N_4, a non-metallic semiconductor, was reported as effective visible light active photo-catalyst since it has small band gap, thermal and chemical stabilities (Masih, 2017; Ragupathi, 2020). The synthesis of g-C_3N_4 based nanocomposites especially with graphene improves the poor conductivity of the g-C_3N_4 (Zhang, 2011; Li, 2013). It is reported that the band gap, conduction band (CB) edge potential and thus the valance band (VB) edge potential of g-C_3N_4 can be tuned effectively by intercalation of various amounts of the rGO (Zhang, 2011; Li, 2013).

In this work, g-C_3N_4 / 3% rGO nanocomposite is synthesized in order to use it as the photo-electrode in the Li-ion oxygen battery. The aim of our work was to get improvement in the discharge-charge performance of the Li-ion oxygen battery by the photo-assistance.

Method

Graphene oxide (GO) was synthesized by the method reported in our previous work (Çelikbilek, 2022). The nanocomposite was synthesized by mixing the melamine and rGO in ethanol at 50°C until all the methanol evaporates. After drying, the mixture was put into a crucible and heated up to 550°C at a rate of 3°C min^{-1} and then kept at this temperature for another 3 h under continues Ar flow, subsequently cooled to room temperature.

Photo-current measurements were made by linear sweep voltammetry technique in a conventional three-electrode cell with a platinum wire as the auxiliary electrode and Ag/AgCl (saturated KCl) as the reference electrode on a Gamry Reference 3000 workstation. A solar simulator (A-type 150 W, 1-3 SUN, Xenon lamb, AMO filters; 400 - 700 nm wavelength) was used as the light source. Measurements were conducted in a spectral cell contains 0.1 M KCl buffered by 0.1 M K_2HPO_4 to pH 7. For the photo-assisted charge-discharge tests the cell was assembled in an Ar-filled glove box with H_2O and O_2 levels less than 0.1 ppm. Lithium metal was used as both counter and reference electrodes and the glass microfiber filter (Whatman) was used as a separator. rGO/Super P carbon black/PVDF were mixed (80:10:10 wt%) in NMP and the slurry was coated onto one side of 16 mm - diameter GDL (TGP-H-060) with a loading rate of 0.1 mg cm^{-2} as a cathode.

X-ray diffraction (XRD) analyses were performed on a PANalytical Empyrean diffractometer with Cu K-alpha radiation at a scanning rate of 2° min^{-1}. The morphologies were examined with a ZEISS Ultraplus scanning electron microscope (SEM). UV/vis spectra were gathered by Cary 5000 UV/Vis/NIR spectrometer with diffuse reflectance accessory between 200 nm and 800 nm.

Results and Discussion

Structure and Morphology

The morphology of the synthesized nanocomposite is shown in Figure 1. The nanocomposite has slate-like stacked lamellar microstructure. Figure 2 shows the XRD patterns of the pure g-C_3N_4, pure rGO and the synthesized nanocomposite. The broad peak located at around 26° in the rGO pattern is ascribed to the presence of the loosely stacked sheets. A strong characteristic (002) peak at 27.6° in the pure g-C_3N_4 pattern is also indication of the layered structure. Another characteristic peak (100) at around 13.2° in the pure g-C_3N_4 pattern in Figure 2 corresponds to the in-plane ordering of tri-s-triazine units. The nanocomposite has almost the same characteristic peaks with the pure g-C_3N_4 in Figure 2.

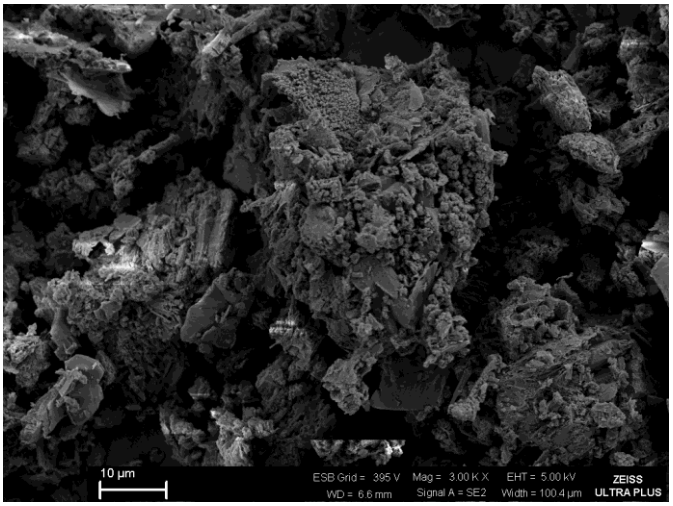

Figure 1. Morphology of the synthesized nanocomposite.

Optical Properties

The photo-anodic currents of pure g-C_3N_4, g-C_3N_4 / 3% rGO nanocomposite and g-C_3N_4 / 5% rGO nanocomposite are provided in Figure 3. The g-C_3N_4 / 3% rGO nanocomposite has improved photo-catalytic efficiency. The enhancement in the photo-currents with the presence of rGO can be attributed to the efficient visible light utilization. The further increase in the rGO content (g-C_3N_4 / 5% rGO), however, degrades this utilization and the photo-currents decline due to the more incident light absorption by rGO instead of g-C_3N_4 in Figure 3.

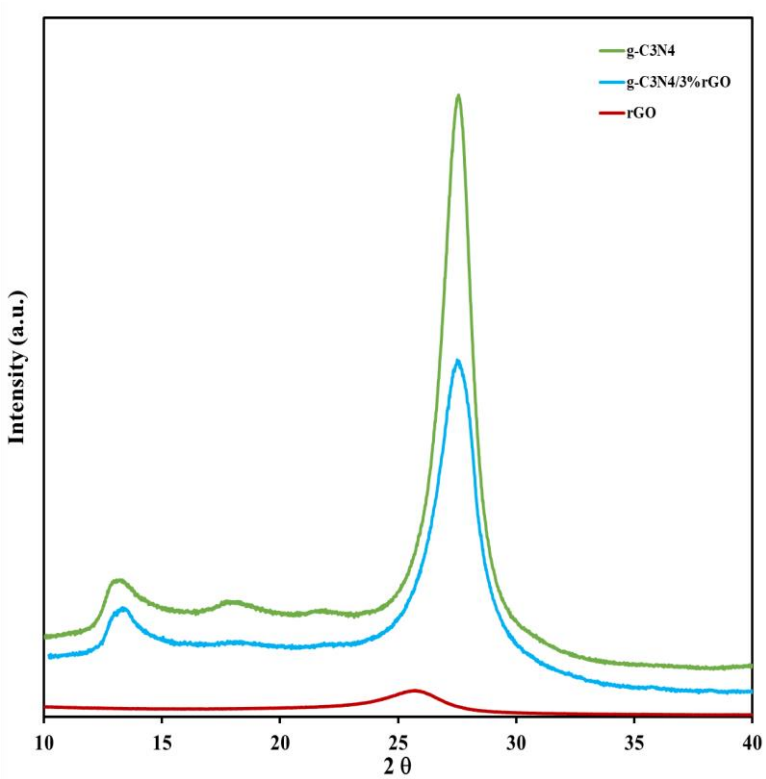

Figure 2. The XRD patterns the pure g-C_3N_4, pure rGO and synthesized nanocomposite.

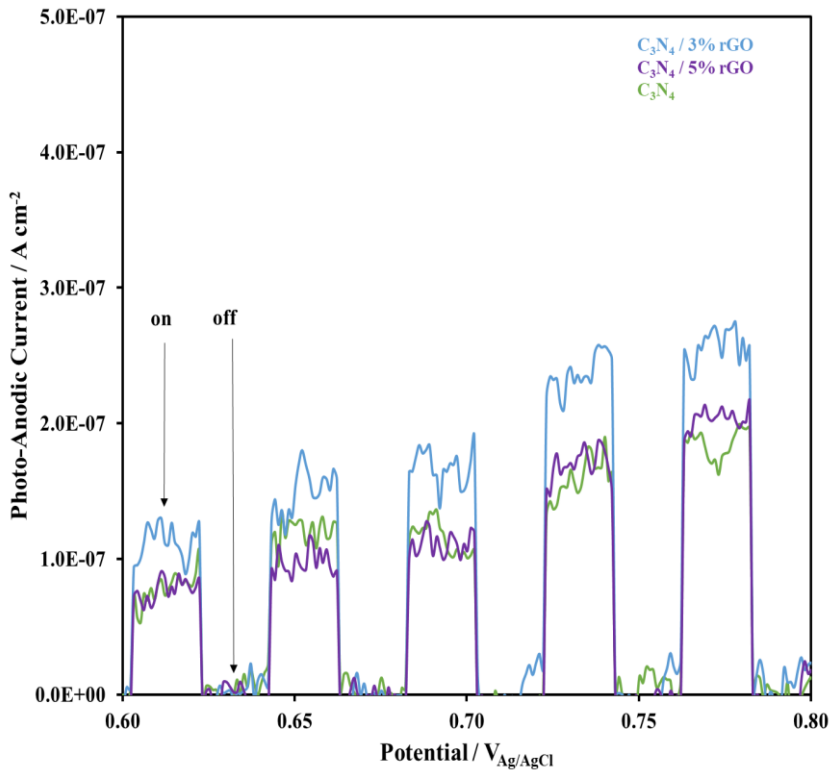

Figure 3. The photo-anodic currents of pure g-C_3N_4, g-C_3N_4 / 3% rGO and g-C_3N_4 / 5% rGO nanocomposites.

The optical band gaps (Eg) obtained from the UV-Vis diffuse reflectance spectra for the pure g-C_3N_4 and nanocomposites are given in Figure 4 that they are 2.7 eV, 2.5 eV and 2.25 eV for g-C_3N_4, g-C_3N_4 / 3% rGO and g-C_3N_4 / 5% rGO, respectively. The narrowing in the optical band gaps depend on the increase in the rGO content of the nanocomposites is attributed to a red shift in the absorption band edge (Li, 2013).

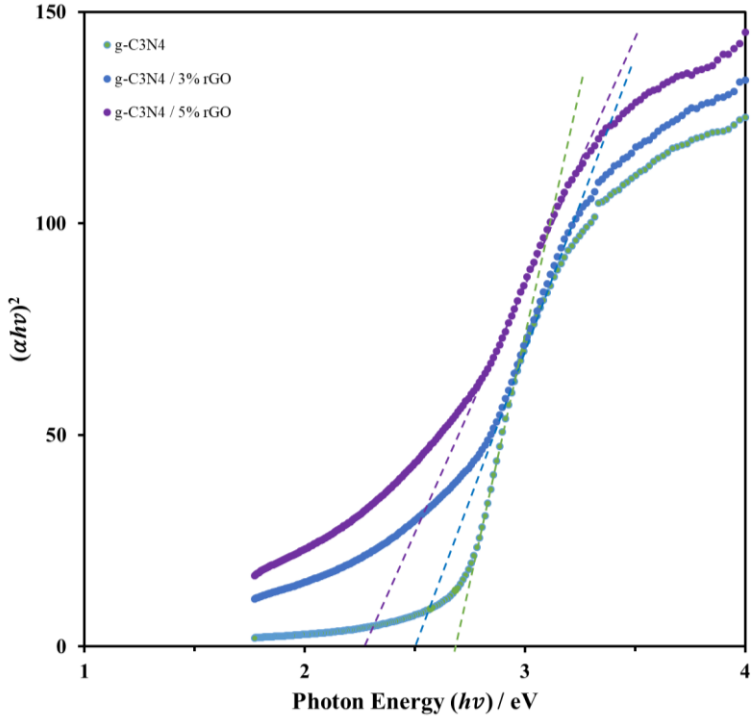

Figure 4. The optical band gaps of g-C_3N_4, g-C_3N_4 / 3% rGO and g-C_3N_4 / 5% rGO nanocomposites.

Photo-Assisted Charging of Li-Ion Oxygen Battery

The photo-assisted discharge-charge performance of the Li-ion oxygen battery for a 0.25 mA h cm^{-2} constant capacity at 0.02 mA cm^{-2} current density is shown by providing 1st, 10th and 50th cycle curves in Figure 5. Obviously, the charge potentials remain under 3.5 $V_{Li+/Li}$ with the photo-assistance for 50 cycles that the Li-ion oxygen battery performance seems improved significantly.

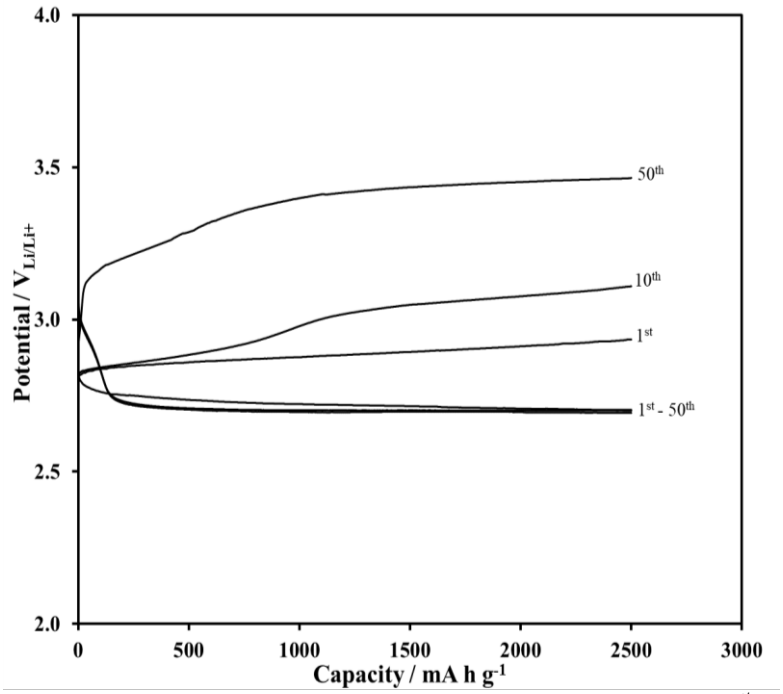

Figure 5. The photo-assisted discharge-charge curves of the Li-ion oxygen battery for 1st, 10th and 50th cycles.

Conclusion

g-C_3N_4 / 3% rGO nanocomposite was synthesized as the photo-catalyst for the photo-assisted charging of the Li-ion oxygen battery. Optical characterizations showed that there was a reduction in the optical band gap of the nanocomposite as a result of the red shift. The usage of this nanocomposite as the photo-electrode in the Li-ion oxygen battery resulted in a considerable reduction in the charge potential and improved the battery cyclic performance.

Scientific Ethics Declaration

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

Acknowledgements or Notes

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