RESEARCH PAPER

Structural and Optical Properties of Graphene/Chlorophyll Pigment Nanocomposite

*Elaf Ayad Khadim *, Mohammed H. K. AL-Mamoori, Saif Mohammed Hassan*

Department of physics, College of Science for Women, University of Babylon, Hilla, Iraq

ARTICLE INFO

Article History:
Received 09 April 2025
Accepted 23 June 2025
Published 01 July 2025

Keywords:
Graphene/Chlorophyll
Laser ablation
Nanocomposite

ABSTRACT

Compared with their bulky or nanosized counterparts, nanostructured carbon materials attract many researchers due to their diverse practical applications. Graphene has been successfully fabricated using a pulsed laser ablation process. It relies on the exfoliation of a graphite disc to induce polycarbonate sheets suspended in an aqueous medium (deionized water). Chlorophyll was extracted from spinach and then mixed with graphene. The properties were studied using a scanning electron microscope (SEM) and X-ray diffraction (XRD). Various graphene nanostructures have been observed, such as disks, dots, and fibers. The optical properties were studied by UV-vis spectroscopy and photoluminescence (PL) measurements. PL of graphene typically shows strong photoemission in the visible region (400.7 nm, 675.3 nm).

How to cite this article
Khadim E., AL-Mamoori M., Hassan S. Structural and Optical Properties of Graphene/Chlorophyll Pigment Nanocomposite. J Nanostruct, 2025; 15(3):1034-1040. DOI: 10.22052/JNS.2025.03.020

INTRODUCTION

Graphene is known as an atomic layer of graphite, which is also the essential unit for fullerenes and CNTs. It is a two dimensional (2D) crystal that is stable under ambient conditions [1, 2]. Graphene has exceptional in-plane structural, mechanical, thermal and electrical properties. These properties make it attractive for application in many research fields [3, 4]. Optoelectronic devices based on graphene have garnered significant interest and show potential for use in solar cells, touch screens, and photodetectors [5-10]. The exceptional optical characteristics of graphene, such as linear optical absorption [11, 12], tunable band-gap [13], and intrinsic photocurrent have been established [14-17]. These qualities can be combined with other unique mechanical and electrical characteristics of graphene to provide novel functions. The weakness of pristine graphene's absorption, which can reach 2.3%, severely restricts its use in photodetection.

As a result, other strategies have been used to improve the interaction between graphene and photons, such as plasmonic effects [18, 19], photothermoelectric effects [20], sensitization by quantum dots [10, 21, 22], or bulk semiconductors [23]. In this work, we employ chlorophyll as a light-absorbing material for graphene phototransistors and examine the hybrid graphene-chlorophyll devices' optoelectronic performance. As an organic semiconductor, chlorophyll absorbs light very well. Furthermore, chlorophyll is an exceptionally stable and abundant biomaterial [24]. Studies on chlorophyll photosensitization could be useful for organic photovoltaics [25, 26], optical sensors [27], and artificial photosynthesis [28], since chlorophyll

* Corresponding Author Email: elaf.ayad1991@gmail.com

is essential to the process of photosynthesis. Furthermore, because chlorophyll-related molecules come in a variety of forms, it is possible to tailor the energy level alignment in hybrid graphene-organic molecule systems to maximize charge separation and transfer. There are two types of chlorophyll—types A and B—found in green algae and terrestrial plants. The presence of methyl in chlorophyll a, which is substituted by a formyl group in chlorophyll b, is what distinguishes these two types of chlorophylls. In higher plants, the ratio of chlorophyll a to chlorophyll b is roughly 3:1. The visible spectrum's red (650–700 nm) and blue–violet (400–500 nm) bands are the primary wavelengths that chlorophyll absorbs [29].

MATERIALS AND METHODS

Pulse laser ablation

Graphene powders can be obtained by using Q-Switched Nd: YAG pulsed laser in liquids as showed in Fig. 1.

Preparation of chlorophyll (collection of plants and chlorophyll extraction)

The spinach leaves from the plant is collected. Ten milliliters of 80% acetone were used to grind

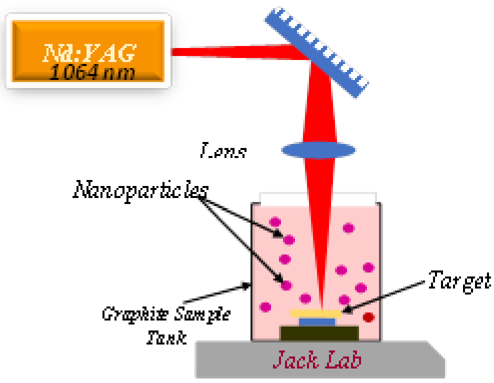

Fig. 1. Schematic diagram of PLAL method to produce graphene.

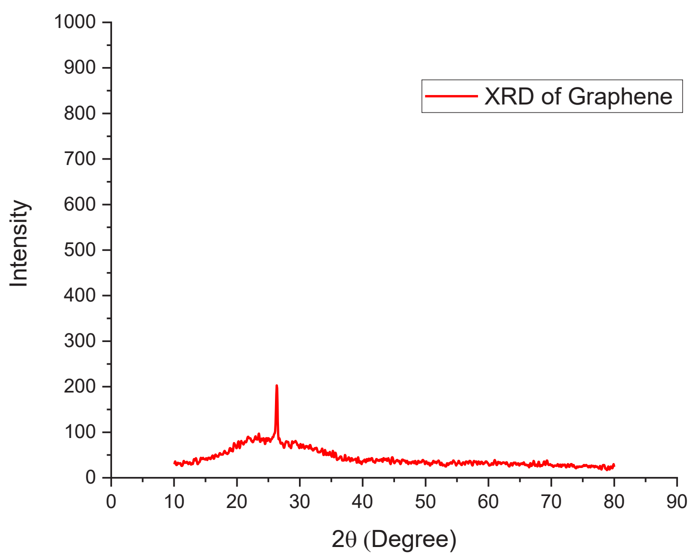

Fig. 2. XRD pattern of graphene.

five grams of freshly chopped leaves. Then, for five minutes, it was centrifuged at 5000–10000 rpm. Once the supernatant was moved, the process was repeated until the residue lost all of its color [29].

RESULTS AND DISCUSSION

XRD pattern

A significant peak at 2θ = 26° can be seen in the XRD pattern of graphene, as illustrated in Fig. 2. It's a unique peak that appear to be quite broad with lesser intensity when compared to bulk graphite's patterns, which is consistent with the previous report [30].

Morphological study

Using FESEM, the surface morphology was examined. The graphene nanoparticles have a

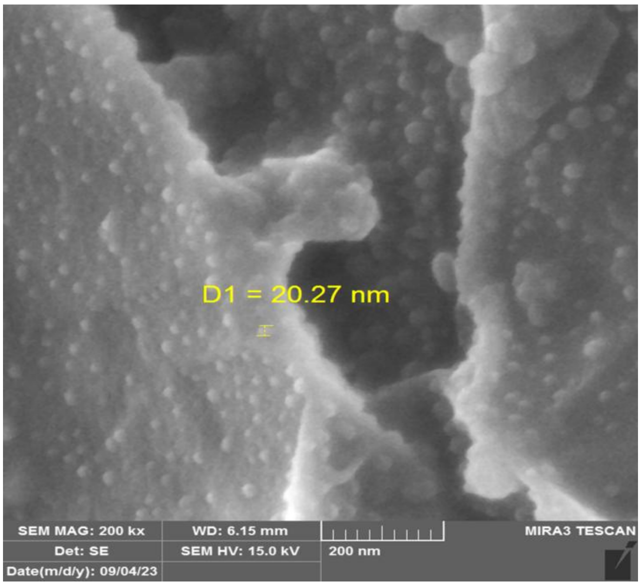

Fig. 3. FE-SEM image of graphene powders.

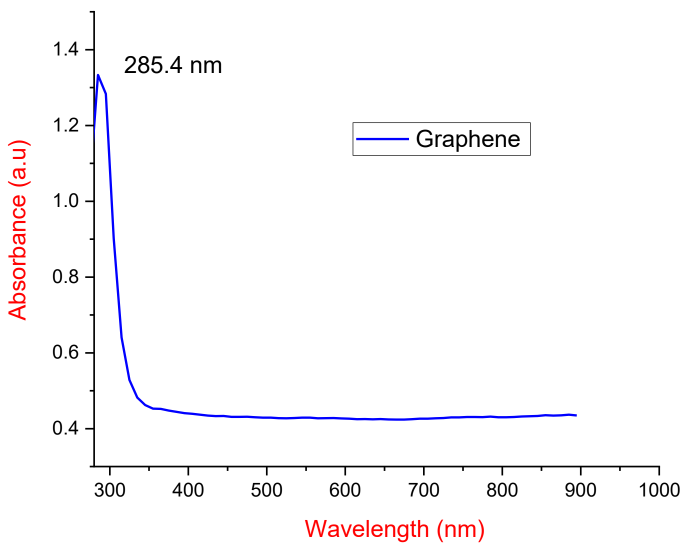

Fig. 4. UV-Vis spectrum of graphene.

spherical shape and different sizes and have an almost homogeneous distribution as showed in Fig. 3. The Image J program was used to calculate the average particle size and it was about 40 nm, which is close to what the researcher reported in the reference [31].

Optical properties

UV-visible spectrum

The UV-visible spectrum of graphene is displayed in Fig. 4. According to earlier research, graphene has an absorption peak at 285.4nm [32]. The electrical arrangement of graphene during the reduction of graphene oxide is the cause of this peak. The n-π* transition of C–O bonds, which are now embedded on the graphene due to exfoliation and intercalation, is responsible for this absorption peak.

Fig. 5 illustrates the UV-Vis spectrum of chlorophyll extract dissolved in acetone

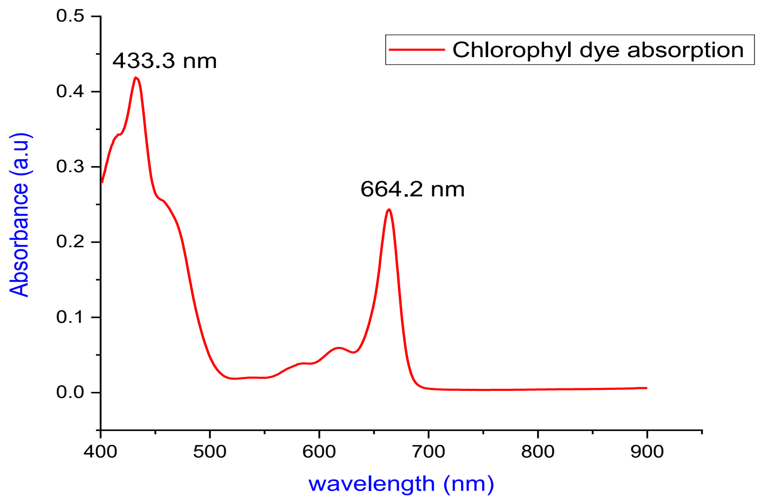

Fig. 5. UV-Vis spectrum of chlorophyll.

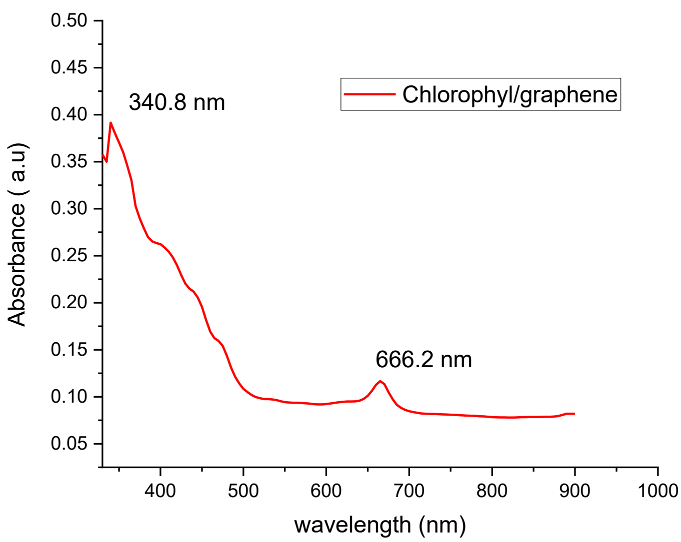

Fig. 6. UV-Vis spectrum of graphene/chlorophyll sample.

absorption. The highest absorption was seen at 433.3 and 664.2 nm [33].

Fig. 6 shows the UV-Vis spectrum of graphene/chlorophyll sample. The absorption peaks for the chlorophyll/graphene are located at 340.8 and 666.2 nm.

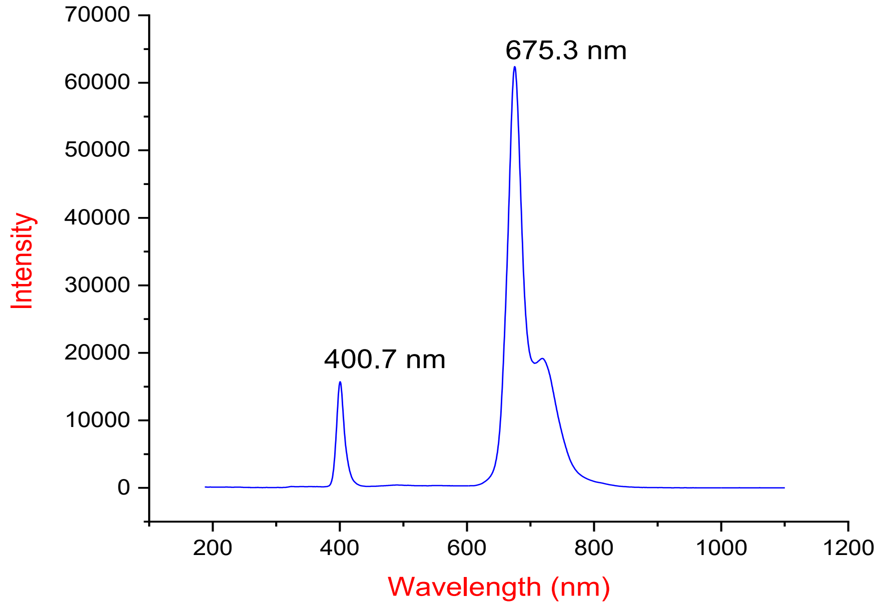

Fig. 7. PL spectrum of graphene nanoparticales.

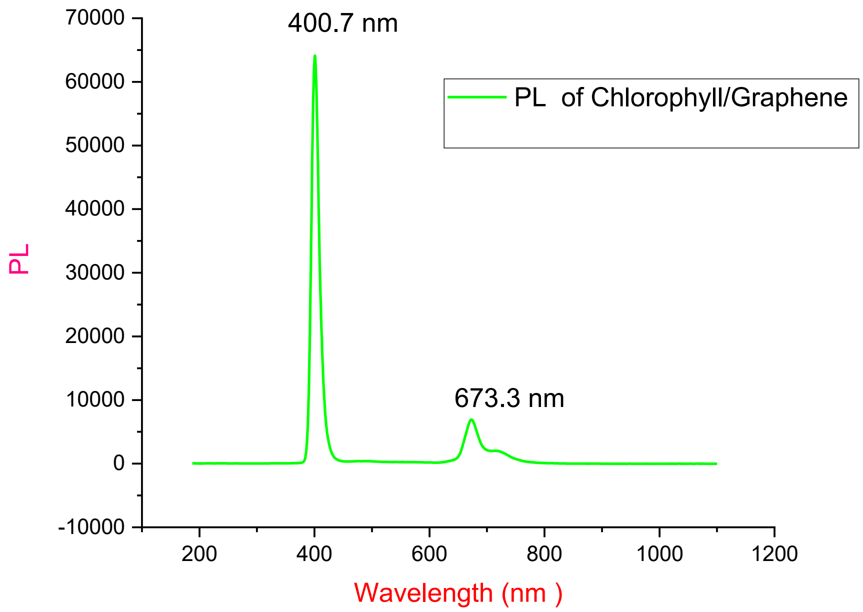

Fig. 8. PL spectrum of graphene/chlorophyll sample.

Photoluminescence

The researchers [34] affirmed that the emission peak of carbon is usually wide with large stocks shift as compared with that of organic dyes and it may result from the wide distributions of dierently sized particles and surface chemistry, different emissive traps (salvation effect), or a mechanism currently unresolved [35]. The emission of the graphene typically shows wide optical emission in the visible region (Fig. 7).

Also, the additional PL data of graphene/chlorophyll sample is shown in Fig. 8. In this sample, there is a clear quenching of PL. The PL quenching suggests that charge transfer occurs between the chlorophyll film and graphene, which reduces radiative recombination since electron–hole pairs are produced in the chlorophyll molecules under illumination [36].

CONCLUSION

The pulsed laser, with a wavelength of 1064 nm, energy of 80 mJ, a frequency of 6 Hz, and a number of pulses of 400 pulses, has the ability to generate nanoparticles having a spherical shape and homogeneous distribution with absorption peak at 285.4nm. The pulsed laser has the ability to exfoliate graphite layers through the appearance of a single peak of low intensity as compared to graphite. Through the PL results, we notice that mixing chlorophyll with graphene nanoparticles enhanced the emission spectrum at 400.7 nm, and this indicates the removal of a small portion of impurities during the mixing process and showed quenching of graphene as a result of charge transfer from chlorophyll, which caused a reduction in radiative recombination.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this manuscript.

REFERENCES

1. Deng J, You Y, Sahajwalla V, Joshi RK. Transforming waste into carbon-based nanomaterials. Carbon. 2016;96:105-115.
2. Terzopoulou Z, Kyzas GZ, Bikiaris DN. Recent Advances in Nanocomposite Materials of Graphene Derivatives with Polysaccharides. Materials (Basel, Switzerland). 2015;8(2):652-683.
3. McAllister MJ, Li J-L, Adamson DH, Schniepp HC, Abdala AA, Liu J, et al. Single Sheet Functionalized Graphene by Oxidation and Thermal Expansion of Graphite. Chem Mater. 2007;19(18):4396-4404.
4. Stankovich S, Dikin DA, Piner RD, Kohlhaas KA, Kleinhammes A, Jia Y, et al. Synthesis of graphene-based nanosheets via chemical reduction of exfoliated graphite oxide. Carbon. 2007;45(7):1558-1565.
5. Lü M, Li J, Yang X, Zhang C, Yang J, Hu H, et al. Applications of graphene-based materials in environmental protection and detection. Chin Sci Bull. 2013;58(22):2698-2710.
6. Bonaccorso F. Solution processed graphene for photonics and optoelectronics. Nonlinear Optics: OSA; 2013. p. NTh1A.1.
7. Bao Q, Loh KP. Graphene Photonics, Plasmonics, and Broadband Optoelectronic Devices. ACS Nano. 2012;6(5):3677-3694.
8. Avouris P, Xia F. Graphene applications in electronics and photonics. MRS Bull. 2012;37(12):1225-1234.
9. Miao X, Tongay S, Petterson MK, Berke K, Rinzler AG, Appleton BR, et al. High Efficiency Graphene Solar Cells by Chemical Doping. Nano Lett. 2012;12(6):2745-2750.
10. Konstantatos G, Badioli M, Gaudreau L, Osmond J, Bernechea M, de Arquer FPG, et al. Hybrid graphene–quantum dot phototransistors with ultrahigh gain. Nature Nanotechnology. 2012;7(6):363-368.
11. Nair RR, Blake P, Grigorenko AN, Novoselov KS, Booth TJ, Stauber T, et al. Fine Structure Constant Defines Visual Transparency of Graphene. Science. 2008;320(5881):1308-1308.
12. Casiraghi C, Hartschuh A, Lidorikis E, Qian H, Harutyunyan H, Gokus T, et al. Rayleigh Imaging of Graphene and Graphene Layers. Nano Lett. 2007;7(9):2711-2717.
13. Zhang Y, Tang T-T, Girit C, Hao Z, Martin MC, Zettl A, et al. Direct observation of a widely tunable bandgap in bilayer graphene. Nature. 2009;459(7248):820-823.
14. Balasubramanian K, Lee EJH, Weitz RT, Burghard M, Kern K. Carbon Nanotube Transistors– Chemical Functionalisation and Device Characterisation. Organic Electronics: Wiley-VCH Verlag GmbH & Co. KGaA. p. 565-593.
15. Mueller T, Xia F, Freitag M, Tsang J, Avouris P. Role of contacts in graphene transistors: A scanning photocurrent study. Physical Review B. 2009;79(24).
16. Xu X, Gabor NM, Alden JS, van der Zande AM, McEuen PL. Photo-Thermoelectric Effect at a Graphene Interface Junction. Nano Lett. 2009;10(2):562-566.
17. Freitag M, Low T, Xia F, Avouris P. Photoconductivity of biased graphene. Nature Photonics. 2012;7(1):53-59.
18. Echtermeyer TJ, Britnell L, Jasnos PK, Lombardo A, Gorbachev RV, Grigorenko AN, et al. Strong plasmonic enhancement of photovoltage in graphene. Nature Communications. 2011;2(1).
19. de Abajo FJGa, Koppens FHL, Chang DE, Thongrattanasiri S, Chigrin DN. Graphene Plasmonics. AIP Conference Proceedings: AIP; 2011. p. 28-29.
20. Ma Q, Gabor NM, Andersen TI, Nair NL, Watanabe K, Taniguchi T, et al. Competing Channels for Hot-Electron Cooling in Graphene. Phys Rev Lett. 2014;112(24).
21. Sun Z, Liu Z, Li J, Tai Ga, Lau SP, Yan F. Infrared Photodetectors Based on CVD-Grown Graphene and PbS Quantum Dots with Ultrahigh Responsivity. Adv Mater. 2012;24(43):5878-5883.
22. Zhang D, Gan L, Cao Y, Wang Q, Qi L, Guo X. Understanding Charge Transfer at PbS-Decorated Graphene Surfaces toward a Tunable Photosensor. Adv Mater. 2012;24(20):2715-2720.
23. Li X, Fan L, Li Z, Wang K, Zhong M, Wei J, et al. Boron Doping of Graphene for Graphene–Silicon p–n Junction Solar Cells. Advanced Energy Materials. 2012;2(4):425-429.
24. Gowda S, Mathur G, Qihang L, Surthi S, Qian Z, Lindsey JS, et al. Hybrid silicon/molecular memories: co-engineering

for novel functionality. IEEE International Electron Devices Meeting 2003: IEEE. p. 22.21.21-22.21.24.
25. Durrant J. Towards optimisation of electron transfer processes in dye sensitised solar cells. Coord Chem Rev. 2004.
26. Spanggaard H. A brief history of the development of organic and polymeric photovoltaics. Sol Energy Mater Sol Cells. 2004;83(2-3):125-146.
27. Hecht DS, Ramirez RJA, Briman M, Artukovic E, Chichak KS, Stoddart JF, et al. Bioinspired Detection of Light Using a Porphyrin-Sensitized Single-Wall Nanotube Field Effect Transistor. Nano Lett. 2006;6(9):2031-2036.
28. Yamada H, Imahori H, Nishimura Y, Yamazaki I, Fukuzumi S. Enhancement of Photocurrent Generation by ITO Electrodes Modified Chemically with Self-Assembled Monolayers of Porphyrin–Fullerene Dyads. Adv Mater. 2002;14(12):892.
29. Kumari M, Sharma OP, Bagri RK, Nathawat BDS. Management of wilt disease of lentil through bio control agents and organic amendments in Rajasthan. Journal of Pharmacognosy and Phytochemistry. 2020;9(5):3248-3252.
30. Graphene Mass Production Comes Closer with Bulk Wet Chemical Exfoliation of Graphite. Highlights: National Nanomanufacturing Network.
31. Subramanya B, Bhat DK. Novel one-pot green synthesis of graphene in aqueous medium under microwave irradiation using a regenerative catalyst and the study of its electrochemical properties. New J Chem. 2015;39(1):420-430.
32. Dikio ED, Thema FT, Farah AM, Shooto ND. One-step reduction, characterization and magnetic behaviour of exfoliated graphene oxide. Materials Science-Poland. 2013;31(1):59-64.
33. Orona-Navar A, Aguilar-Hernández I, Cerdán-Pasarán A, López-Luke T, Rodríguez-Delgado M, Cárdenas-Chávez DL, et al. Astaxanthin from Haematococcus pluvialis as a natural photosensitizer for dye-sensitized solar cell. Algal Research. 2017;26:15-24.
34. Liu W, Speranza G. Tuning the Oxygen Content of Reduced Graphene Oxide and Effects on Its Properties. ACS omega. 2021;6(9):6195-6205.
35. Saha A, Basiruddin SK, Sarkar R, Pradhan N, Jana NR. Functionalized Plasmonic−Fluorescent Nanoparticles for Imaging and Detection. The Journal of Physical Chemistry C. 2009;113(43):18492-18498.
36. Wu HC, Huang YC, Ding IK, Chen CC, Yang YH, Tsai CC, et al. Photoinduced Electron Transfer in Dye-Sensitized SnO_2 Nanowire Field-Effect Transistors. Adv Funct Mater. 2010;21(3):474-479.