SUPPORTING INFORMATION

For

Graphitic carbon nitride (g-C_3N_4) electrodes for energy conversion and storage: A review on photoelectrochemical water splitting, solar cells and supercapacitors

Javad Safaei[a]*, *Nurul Aida Mohamed*[a], *Mohamad Firdaus Mohamad Noh*[a], *Wan Nor Roslam Wan Isahak*[b], *Mohd Asri Mat Teridi*[a]*

[a] Solar Energy Research Institute, National University of Malaysia, 43600 Bangi, Selangor, Malaysia

[b] Department of Chemical and Process Engineering, Faculty of Engineering and Built Environment, National University of Malaysia, 43600 Bangi, Selangor, Malaysia

Author information

*Corresponding Authors' Email: javad.safaei@outlook.com (*Javad Safaei*), asri@ukm.edu.my (*Mohd Asri Mat Teridi*)

(1) Calculation of maximum theoretical value of photocurrent:

According to Bian et al. the conventional g-C_3N_4 with the band gap of 2.7 eV corresponds to the maximum photocurrent of 4 mAcm^{-2}.[1] The photocurrent density at 1.23 V vs. RHE was calculated via the area of solar spectrum absorption and the obtained value was found to be 4.27 mAcm^{-2} that is approximately identical with the reported value. AM 1.5G solar spectrum was obtained from ASTM.[2] The amount of power absorption was calculated via integration of AM 1.5 G solar spectrum from 300 nm, i.e. based on lower wavelength limit of solar absorption, up to 459 nm, i.e. based on band gap of g-C_3N_4 $\frac{1240}{2.7} = 459\ nm$, according to Equation S1.[3] The IPCE and faradaic efficiency is considered 100% when calculating the amount of STH and maximum photocurrent density.[4] The area of AM 1.5G was calculated and obtained via Origin software (Figure S1).

$$P_{in} = \int_{512}^{533} AM1.5G(E)dE \qquad \textbf{(Equation S1)}$$

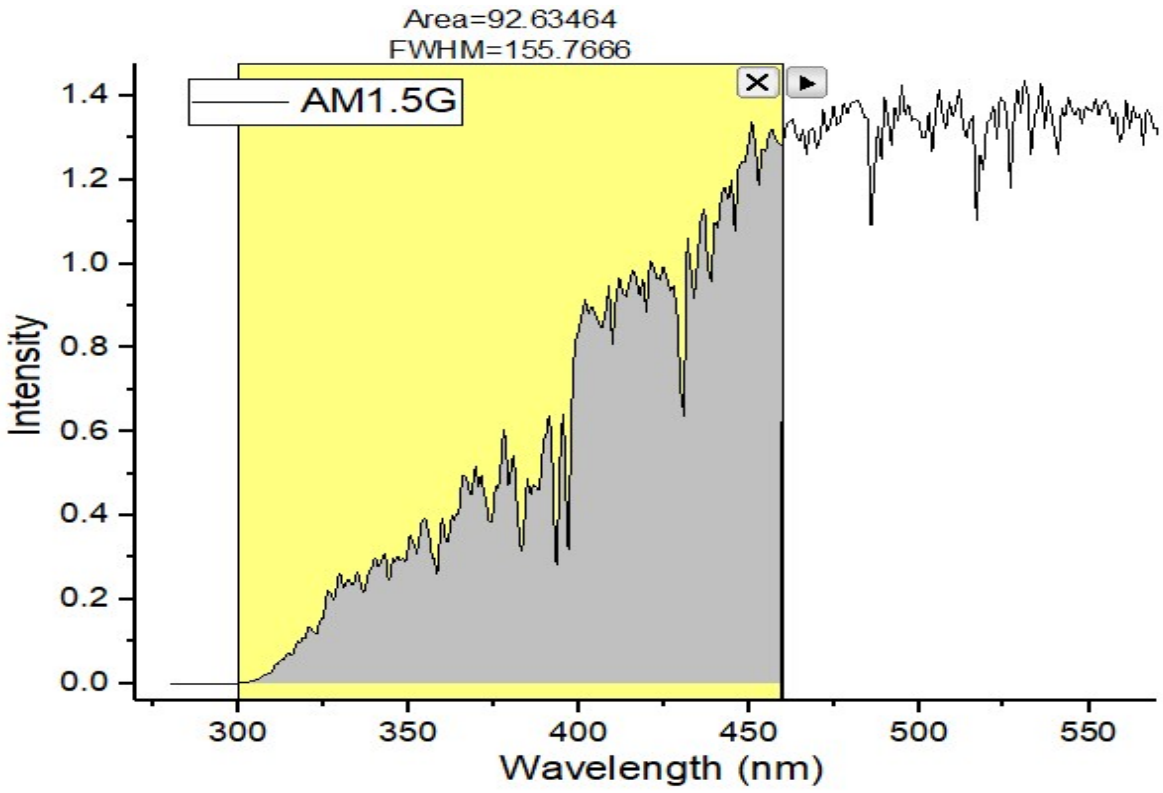

Figure S1: Light absorption for g-C_3N_4 with band gap of 2.7 eV.

This demonstrates the fundamental theoretical and experimental research that is still required to be conducted on g-C_3N_4 photoelectrodes. In fact, extending the absorption edge by reducing the band gap of g-C_3N_4, increases this theoretical value. However, development of photoelectrodes based on g-C_3N_4 (E_g=2.7 eV) with the photocurrent value of nearly 4 mAcm^{-2} should be prioritized in future investigations.

References:

(1) Bian, J.; Li, Q.; Huang, C.; Li, J.; Guo, Y.; Zaw, M.; Zhang, R.-Q. Thermal vapor condensation of uniform graphitic carbon nitride films with remarkable photocurrent density for photoelectrochemical applications. *Nano Energy* **2015,** *15*, 353.

(2) Reference Solar Spectral Irradiance: Air Mass 1.5, American Society for Testing and Materials (ASTM) Terrestrial Reference Spectra for Photovoltaic Performance Evaluation (http://rredc.nrel.gov/solar/spectra/am1.5/).

(3) Fountaine, K. T.; Lewerenz, H. J.; Atwater, H. A. Efficiency limits for photoelectrochemical water-splitting. *Nat. Comm.* **2016,** *7*, 13706.

(4) Hu, S.; Xiang, C.; Haussener, S.; Berger, A. D.; Lewis, N. S. An analysis of the optimal band gaps of light absorbers in integrated tandem photoelectrochemical water-splitting systems. *Energy Environ. Sci.* **2013,** *6* (10), 2984.

(2) The entire references with full titles, numbered according to the main article:

References

(1) Huang, J.; Yu, H.; Dai, A.; Wei, Y.; Kang, L. Drylands face potential threat under 2 C global warming target. *Nat. Clim. Change* **2017,** *7* (6), 417.

(2) Tokarska, K. B.; Gillett, N. P. Cumulative carbon emissions budgets consistent with 1.5° C global warming. *Nat. Clim. Change* **2018,** *8* (4), 296.

(3) Gray, H. B. Powering the planet with solar fuel. *Nat. Chem.* **2009,** *1* (1), 7.

(4) Lewis, N. S. Powering the planet. *MRS Bull.* **2007,** *32* (10), 808.

(5) Lewis, N. S.; Nocera, D. G. Powering the planet: Chemical challenges in solar energy utilization. *Proc. Natl. Acad. Sci.* **2006,** *103* (43), 15729.

(6) Shamshiri, M.; Gan, C. K.; Omar, R. Assessment of distribution networks performance considering residential photovoltaic systems with demand response applications. *J. Renew. Sustain. Energy* **2017,** *9* (4), 045502.

(7) Dubal, D. P.; Ayyad, O.; Ruiz, V.; Gomez-Romero, P. Hybrid energy storage: the merging of battery and supercapacitor chemistries. *Chem. Soc. Rev.* **2015,** *44* (7), 1777.

(8) Hou, Y.; Li, J.; Wen, Z.; Cui, S.; Yuan, C.; Chen, J. N-doped graphene/porous g-C3N4 nanosheets supported layered-MoS2 hybrid as robust anode materials for lithium-ion batteries. *Nano Energy* **2014,** *8*, 157.

(9) Zhang, J.; Li, J. Y.; Wang, W. P.; Zhang, X. H.; Tan, X. H.; Chu, W. G.; Guo, Y. G. Microemulsion Assisted Assembly of 3D Porous S/Graphene@ g-C3N4 Hybrid Sponge as Free-Standing Cathodes for High Energy Density Li–S Batteries. *Adv. Energy Mater.* **2018,** *8* (14), 1702839.

(10) Podjaski, F.; Kröger, J.; Lotsch, B. V. Toward an Aqueous Solar Battery: Direct Electrochemical Storage of Solar Energy in Carbon Nitrides. *Adv. Mater.* **2018,** *30* (9), 1705477.

(11) Li, Q.; Liu, Y.; Guo, S.; Zhou, H. Solar energy storage in the rechargeable batteries. *Nano Today* **2017,** *16*, 46.

(12) Kudo, A.; Miseki, Y. Heterogeneous photocatalyst materials for water splitting. *Chem. Soc. Rev.* **2009,** *38* (1), 253.

(13) Safaei, J.; Ullah, H.; Mohamed, N. A.; Noh, M. F. M.; Soh, M. F.; Tahir, A. A.; Ludin, N. A.; Ibrahim, M. A.; Isahak, W. N. R. W.; Teridi, M. A. M. Enhanced photoelectrochemical performance of Z-scheme g-C3N4/BiVO4 photocatalyst. *Appl. Catal. B* **2018,** *234*, 296.

(14) Bin, A. R.; Yusoff, M.; Jang, J. Highly efficient photoelectrochemical water splitting by a hybrid tandem perovskite solar cell. *Chem. Commun.* **2016,** *52* (34), 5824.

(15) Safaei, J.; Mohamed, N. A.; Noh, M. F. M.; Soh, M. F.; Elbreki, A. M.; Ludin, N. A.; Ibrahim, M. A.; Al-Waeli, A. H. A.; Isahak, W. N. R. W.; Teridi, M. A. M. Simultaneous enhancement in light absorption and charge transportation of bismuth vanadate (BiVO4) photoanode via microwave annealing. *Mater. Lett.* **2018** https://doi.org/10.1016/j.matlet.2018.08.143.

(16) Marbán, G.; Valdés-Solís, T. Towards the hydrogen economy? *Int. J. Hydrogen Energy* **2007,** *32* (12), 1625.

(17) Alanne, K.; Cao, S. Zero-energy hydrogen economy (ZEH2E) for buildings and communities including personal mobility. *Renew. Sustain. Energy Rev.* **2017,** *71*, 697.

(18) Wang, P.; Wang, S.; Wang, H.; Wu, Z.; Wang, L. Recent Progress on Photo-Electrocatalytic Reduction of Carbon Dioxide. *Part. Part. Syst. Charact.* **2018,** *35* (1), 1700371.

(19) Shen, M.; Zhang, L.; Shi, J. Converting CO2 into fuels by graphitic carbon nitride-based photocatalysts. *Nanotechnology* **2018,** *29* (41), 412001.

(20) Fang, Y.; Wang, X. Photocatalytic CO 2 conversion by polymeric carbon nitrides. *Chem. Commun.* **2018**.
(21) Choudhary, N.; Islam, M. A.; Kim, J. H.; Ko, T.-J.; Schropp, A.; Hurtado, L.; Weitzman, D.; Zhai, L.; Jung, Y. Two-dimensional transition metal dichalcogenide hybrid materials for energy applications. *Nano Today* **2018,** *19*, 16.
(22) Li, C.; Cao, Q.; Wang, F.; Xiao, Y.; Li, Y.; Delaunay, J.-J.; Zhu, H. Engineering graphene and TMDs based van der Waals heterostructures for photovoltaic and photoelectrochemical solar energy conversion. *Chem. Soc. Rev.* **2018,** *47*, 4981.
(23) Di, J.; Xiong, J.; Li, H.; Liu, Z. Ultrathin 2D Photocatalysts: Electronic-Structure Tailoring, Hybridization, and Applications. *Adv. Mater.* **2018,** *30* (1), 1704548.
(24) Fu, J.; Yu, J.; Jiang, C.; Cheng, B. g-C3N4-Based Heterostructured Photocatalysts. *Adv. Energy Mater.* **2017,** *8* (3), 1701503.
(25) Kumar, S.; Kumar, A.; Bahuguna, A.; Sharma, V.; Krishnan, V. Two-dimensional carbon-based nanocomposites for photocatalytic energy generation and environmental remediation applications. *Beilstein journal of nanotechnology* **2017,** *8*, 1571.
(26) Jin, H.; Guo, C.; Liu, X.; Liu, J.; Vasileff, A.; Jiao, Y.; Zheng, Y.; Qiao, S.-Z. Emerging Two-Dimensional Nanomaterials for Electrocatalysis. *Chem. Rev.* **2018**.
(27) Meng, X.; Zhang, Z. Two dimensional graphitic materials for photoelectrocatalysis: A short review. *Catal. Today* **2018. DOI: https://doi.org/10.1016/j.cattod.2018.03.015.**
(28) Wang, X.; Maeda, K.; Thomas, A.; Takanabe, K.; Xin, G.; Carlsson, J. M.; Domen, K.; Antonietti, M. A metal-free polymeric photocatalyst for hydrogen production from water under visible light. *Nat. Mater.* **2009,** *8* (1), 76.
(29) Cao, S.; Low, J.; Yu, J.; Jaroniec, M. Polymeric photocatalysts based on graphitic carbon nitride. *Adv. Mater.* **2015,** *27* (13), 2150.
(30) Naseri, A.; Samadi, M.; Pourjavadi, A.; Moshfegh, A. Z.; Ramakrishna, S. Graphitic carbon nitride (gC 3 N 4)-based photocatalysts for solar hydrogen generation: recent advances and future development directions. *J. Mater. Chem. A* **2017,** *5* (45), 23406.
(31) Zhao, Z.; Sun, Y.; Dong, F. Graphitic carbon nitride based nanocomposites: a review. *Nanoscale* **2015,** *7* (1), 15.
(32) Gholipour, M. R.; Dinh, C.-T.; Béland, F.; Do, T.-O. Nanocomposite heterojunctions as sunlight-driven photocatalysts for hydrogen production from water splitting. *Nanoscale* **2015,** *7* (18), 8187.
(33) Zhou, L.; Zhang, H.; Sun, H.; Liu, S.; Tade, M. O.; Wang, S.; Jin, W. Recent advances in non-metal modification of graphitic carbon nitride for photocatalysis: a historic review. *Catal. Sci. Technol.* **2016,** *6* (19), 7002.
(34) Wen, J.; Xie, J.; Chen, X.; Li, X. A review on g-C3N4-based photocatalysts. *Appl. Surf. Sci.* **2017,** *391*, 72.
(35) Liu, J.; Wang, H.; Antonietti, M. Graphitic carbon nitride “reloaded”: emerging applications beyond (photo) catalysis. *Chem. Soc. Rev.* **2016,** *45* (8), 2308.
(36) Zheng, Y.; Lin, L.; Wang, B.; Wang, X. Graphitic carbon nitride polymers toward sustainable photoredox catalysis. *Angew. Chem. Int. Ed.* **2015,** *54* (44), 12868.
(37) Ong, W.-J.; Tan, L.-L.; Ng, Y. H.; Yong, S.-T.; Chai, S.-P. Graphitic carbon nitride (g-C3N4)-based photocatalysts for artificial photosynthesis and environmental remediation: are we a step closer to achieving sustainability? *Chem. Rev.* **2016,** *116* (12), 7159.
(38) Xiao, M.; Luo, B.; Wang, S.; Wang, L. Solar energy conversion on gC 3 N 4 photocatalyst: Light harvesting, charge separation, and surface kinetics. *J. Energy Chem.* **2018. DOI: 10.1016/j.jechem.2018.02.018.**
(39) Ladva, S. A.; Travis, W.; Quesada-Cabrera, R.; Rosillo-Lopez, M.; Afandi, A.; Li, Y.; Jackman, R. B.; Bear, J. C.; Parkin, I. P.; Blackman, C. Nanoscale, conformal films of graphitic carbon nitride deposited at room temperature: a method for construction of heterojunction devices. *Nanoscale* **2017,** *9* (43), 16586.

(40) Wang, R.; Liu, H.; Fan, Z.; Li, L.; Cai, Y.; Xu, G.; Luo, W.; Yang, B.; Zhou, Y.; Zou, Z. Unconventional gas-based bottom-up, meter-area-scale fabrication of hydrogen-bond free g-CN nanorod arrays and coupling layers with TiO 2 toward high-efficiency photoelectrochemical performance. *Nanoscale* **2018,** *10* (7), 3342.
(41) Safaei, J.; Mohamed, N. A.; Noh, M. F. M.; Soh, M. F.; Riza, M. A.; Mustakim, N. S. M.; Ludin, N. A.; Ibrahim, M. A.; Isahak, W. N. R. W.; Teridi, M. A. M. Facile fabrication of graphitic carbon nitride,(g-C3N4) thin film. *J. Alloys Compd.* **2018,** *769*, 130.
(42) Volokh, M.; Peng, G.; Barrio, J.; Shalom, M. Carbon Nitride Materials for Water Splitting Photoelectrochemical Cells. *Angew. Chem. Int. Ed.* **2018. DOI:**

10.1002/ange.201806514.
(43) Marschall, R. Semiconductor composites: strategies for enhancing charge carrier separation to improve photocatalytic activity. *Adv. Funct. Mater.* **2014,** *24* (17), 2421.
(44) Nellist, M. R.; Laskowski, F. A.; Lin, F.; Mills, T. J.; Boettcher, S. W. Semiconductor–electrocatalyst interfaces: theory, experiment, and applications in photoelectrochemical water splitting. *Acc. Chem. Res.* **2016,** *49* (4), 733.
(45) Hisatomi, T.; Kubota, J.; Domen, K. Recent advances in semiconductors for photocatalytic and photoelectrochemical water splitting. *Chem. Soc. Rev.* **2014,** *43* (22), 7520.
(46) Wang, C.-H.; Qin, D.-D.; Shan, D.-L.; Gu, J.; Yan, Y.; Chen, J.; Wang, Q.-H.; He, C.-H.; Li, Y.; Quan, J.-J. Assembly of gC 3 N 4-based type II and Z-scheme heterojunction anodes with improved charge separation for photoelectrojunction water oxidation. *Phys. Chem. Chem. Phys.* **2017,** *19* (6), 4507.
(47) Hu, S.; Shaner, M. R.; Beardslee, J. A.; Lichterman, M.; Brunschwig, B. S.; Lewis, N. S. Amorphous TiO2 coatings stabilize Si, GaAs, and GaP photoanodes for efficient water oxidation. *Science* **2014,** *344* (6187), 1005.
(48) Wang, X.; Peng, K.-Q.; Hu, Y.; Zhang, F.-Q.; Hu, B.; Li, L.; Wang, M.; Meng, X.-M.; Lee, S.-T. Silicon/hematite core/shell nanowire array decorated with gold nanoparticles for unbiased solar water oxidation. *Nano Lett.* **2013,** *14* (1), 18.
(49) AlOtaibi, B.; Fan, S.; Vanka, S.; Kibria, M.; Mi, Z. A metal-nitride nanowire dual-photoelectrode device for unassisted solar-to-hydrogen conversion under parallel illumination. *Nano Lett.* **2015,** *15* (10), 6821.
(50) Young, J. L.; Steiner, M. A.; Döscher, H.; France, R. M.; Turner, J. A.; Deutsch, T. G. Direct solar-to-hydrogen conversion via inverted metamorphic multi-junction semiconductor architectures. *Nat. Energy* **2017,** *2* (4), 17028.
(51) Barreca, D.; Carraro, G.; Gasparotto, A.; Maccato, C.; Warwick, M. E.; Kaunisto, K.; Sada, C.; Turner, S.; Gönüllü, Y.; Ruoko, T. P. Fe2O3–TiO2 Nano-heterostructure Photoanodes for Highly Efficient Solar Water Oxidation. *Adv. Mater. Interfaces* **2015,** *2* (17), 1500313.
(52) An, X.; Li, T.; Wen, B.; Tang, J.; Hu, Z.; Liu, L. M.; Qu, J.; Huang, C.; Liu, H. New Insights into Defect-Mediated Heterostructures for Photoelectrochemical Water Splitting. *Adv. Energy Mater.* **2016,** *6* (8), 1502268.
(53) Barroso, M.; Pendlebury, S. R.; Cowan, A. J.; Durrant, J. R. Charge carrier trapping, recombination and transfer in hematite (α-Fe 2 O 3) water splitting photoanodes. *Chem. Sci.* **2013,** *4* (7), 2724.
(54) Pendlebury, S. R.; Wang, X.; Le Formal, F.; Cornuz, M.; Kafizas, A.; Tilley, S. D.; Grätzel, M.; Durrant, J. R. Ultrafast charge carrier recombination and trapping in hematite photoanodes under applied bias. *J. Am. Chem. Soc.* **2014,** *136* (28), 9854.
(55) Wang, Y.; Wang, X.; Antonietti, M. Polymeric graphitic carbon nitride as a heterogeneous organocatalyst: from photochemistry to multipurpose catalysis to sustainable chemistry. *Angew. Chem. Int. Ed.* **2012,** *51* (1), 68.
(56) Zhang, S.; Li, J.; Wang, X.; Huang, Y.; Zeng, M.; Xu, J. Rationally designed 1D Ag@ AgVO 3 nanowire/graphene/protonated gC 3 N 4 nanosheet heterojunctions for enhanced

photocatalysis via electrostatic self-assembly and photochemical reduction methods. *J. Mater. Chem. A* **2015,** *3* (18), 10119.
(57) Zhang, J.; Zhang, M.; Sun, R. Q.; Wang, X. A facile band alignment of polymeric carbon nitride semiconductors to construct isotype heterojunctions. *Angew. Chem.* **2012,** *124* (40), 10292.
(58) Zhang, X.; Wang, H.; Wang, H.; Zhang, Q.; Xie, J.; Tian, Y.; Wang, J.; Xie, Y. Single-Layered Graphitic-C3N4 Quantum Dots for Two-Photon Fluorescence Imaging of Cellular Nucleus. *Adv. Mater.* **2014,** *26* (26), 4438.
(59) Lin, Q.; Li, L.; Liang, S.; Liu, M.; Bi, J.; Wu, L. Efficient synthesis of monolayer carbon nitride 2D nanosheet with tunable concentration and enhanced visible-light photocatalytic activities. *Appl. Catal. B* **2015,** *163*, 135.
(60) Tong, J.; Zhang, L.; Li, F.; Wang, K.; Han, L.; Cao, S. Rapid and high-yield production of gC 3 N 4 nanosheets via chemical exfoliation for photocatalytic H 2 evolution. *RSC Adv.* **2015,** *5* (107), 88149.
(61) Niu, P.; Zhang, L.; Liu, G.; Cheng, H. M. Graphene-like carbon nitride nanosheets for improved photocatalytic activities. *Adv. Funct. Mater.* **2012,** *22* (22), 4763.
(62) Han, Q.; Wang, B.; Gao, J.; Cheng, Z.; Zhao, Y.; Zhang, Z.; Qu, L. Atomically thin mesoporous nanomesh of graphitic C3N4 for high-efficiency photocatalytic hydrogen evolution. *ACS Nano* **2016,** *10* (2), 2745.
(63) Di, J.; Xia, J.; Li, H.; Liu, Z. Freestanding atomically-thin two-dimensional materials beyond graphene meeting photocatalysis: Opportunities and challenges. *Nano Energy* **2017,** *35*, 79.
(64) Zhu, S.; Song, Y.; Wang, J.; Wan, H.; Zhang, Y.; Ning, Y.; Yang, B. Photoluminescence mechanism in graphene quantum dots: Quantum confinement effect and surface/edge state. *Nano Today* **2017,** *13*, 10.
(65) Zhang, G.; Zhang, J.; Zhang, M.; Wang, X. Polycondensation of thiourea into carbon nitride semiconductors as visible light photocatalysts. *J. Mater. Chem.* **2012,** *22* (16), 8083.
(66) Dong, F.; Zhao, Z.; Xiong, T.; Ni, Z.; Zhang, W.; Sun, Y.; Ho, W.-K. In situ construction of g-C3N4/g-C3N4 metal-free heterojunction for enhanced visible-light photocatalysis. *ACS Appl. Mater. Interfaces* **2013,** *5* (21), 11392.
(67) Ma, X.; Zhang, J.; Wang, B.; Li, Q.; Chu, S. Hierarchical Cu2O foam/g-C3N4 photocathode for photoelectrochemical hydrogen production. *Appl. Surf. Sci.* **2018,** *427*, 907.
(68) Zhang, J.; Chen, X.; Takanabe, K.; Maeda, K.; Domen, K.; Epping, J. D.; Fu, X.; Antonietti, M.; Wang, X. Synthesis of a carbon nitride structure for visible-light catalysis by copolymerization. *Angew. Chem. Int. Ed.* **2010,** *49* (2), 441.
(69) Zhang, J.; Zhang, M.; Lin, L.; Wang, X. Sol Processing of Conjugated Carbon Nitride Powders for Thin-Film Fabrication. *Angew. Chem.* **2015,** *127* (21), 6395.
(70) Bi, L.; Xu, D.; Zhang, L.; Lin, Y.; Wang, D.; Xie, T. Metal Ni-loaded gC 3 N 4 for enhanced photocatalytic H 2 evolution activity: the change in surface band bending. *Phys. Chem. Chem. Phys.* **2015,** *17* (44), 29899.
(71) Wang, H.; Zhang, X.; Xie, J.; Zhang, J.; Ma, P.; Pan, B.; Xie, Y. Structural distortion in graphitic-C 3 N 4 realizing an efficient photoreactivity. *Nanoscale* **2015,** *7* (12), 5152.
(72) Li, H.; Yu, H.; Quan, X.; Chen, S.; Zhang, Y. Uncovering the key role of the Fermi level of the electron mediator in a Z-scheme photocatalyst by detecting the charge transfer process of WO3-metal-gC3N4 (metal= Cu, Ag, Au). *ACS Appl. Mater. Interfaces* **2016,** *8* (3), 2111.
(73) Kang, Y.; Yang, Y.; Yin, L. C.; Kang, X.; Liu, G.; Cheng, H. M. An Amorphous Carbon Nitride Photocatalyst with Greatly Extended Visible-Light-Responsive Range for Photocatalytic Hydrogen Generation. *Adv. Mater.* **2015,** *27* (31), 4572.
(74) Albero, J.; Barea, E. M.; Xu, J.; Mora-Seró, I.; Garcia, H.; Shalom, M. Toward efficient carbon nitride photoelectrochemical cells: Understanding charge transfer processes. *Adv. Mater. Interfaces* **2017,** *4* (1), 1600265.

(75) Qu, D.; Liu, J.; Miao, X.; Han, M.; Zhang, H.; Cui, Z.; Sun, S.; Kang, Z.; Fan, H.; Sun, Z. Peering into water splitting mechanism of g-C3N4-carbon dots metal-free photocatalyst. *Appl. Catal. B* **2018,** *227*, 418.
(76) Ehrmaier, J.; Domcke, W.; Opalka, D. Mechanism of Photocatalytic Water Oxidation by Graphitic Carbon Nitride. *J. Phys. Chem. Lett* **2018. DOI: 10.1021/acs.jpclett.8b02026.**
(77) Ehrmaier, J.; Karsili, T. N.; Sobolewski, A. L.; Domcke, W. Mechanism of Photocatalytic Water Splitting with Graphitic Carbon Nitride: Photochemistry of the Heptazine–Water Complex. *J. Phys. Chem. A* **2017,** *121* (25), 4754.
(78) Ehrmaier, J.; Janicki, M. J.; Sobolewski, A. L.; Domcke, W. Mechanism of photocatalytic water splitting with triazine-based carbon nitrides: insights from ab initio calculations for the triazine–water complex. *Phys. Chem. Chem. Phys.* **2018,** *20* (21), 14420.
(79) Chen, X.; Zhao, X.; Kong, Z.; Ong, W.-J.; Li, N. Unravelling the electrochemical mechanisms for nitrogen fixation on single transition metal atom embedded in defective graphitic carbon nitride. *J. Mater. Chem. A* **2018. DOI: 10.1039/C8TA06497K.**
(80) Zhang, Y.; Pan, Q.; Chai, G.; Liang, M.; Dong, G.; Zhang, Q.; Qiu, J. Synthesis and luminescence mechanism of multicolor-emitting gC 3 N 4 nanopowders by low temperature thermal condensation of melamine. *Sci. Rep.* **2013,** *3*, 1943.
(81) Chen, X.; Li, N.; Kong, Z.; Ong, W.-J.; Zhao, X. Photocatalytic fixation of nitrogen to ammonia: state-of-the-art advancements and future prospects. *Materials Horizons* **2018,** *5* (1), 9.
(82) Kessler, F. K.; Zheng, Y.; Schwarz, D.; Merschjann, C.; Schnick, W.; Wang, X.; Bojdys, M. J. Functional carbon nitride materials—design strategies for electrochemical devices. *Nat. Rev. Mater.* **2017,** *2* (6), 17030.
(83) Ong, W.-J.; Putri, L. K.; Tan, Y.-C.; Tan, L.-L.; Li, N.; Ng, Y. H.; Wen, X.; Chai, S.-P. Unravelling charge carrier dynamics in protonated gC 3 N 4 interfaced with carbon nanodots as co-catalysts toward enhanced photocatalytic CO 2 reduction: A combined experimental and first-principles DFT study. *Nano Res.* **2017,** *10* (5), 1673.
(84) Kong, Z.; Chen, X.; Ong, W.-J.; Zhao, X.; Li, N. Atomic-level insight into the mechanism of 0D/2D black phosphorus quantum dot/graphitic carbon nitride (BPQD/GCN) metal-free heterojunction for photocatalysis. *Appl. Surf. Sci.* **2018,** *463*, 1148.
(85) Liu, X.; Karsili, T. N.; Sobolewski, A. L.; Domcke, W. Photocatalytic water splitting with the acridine chromophore: a computational study. *J. Phys. Chem. B* **2015,** *119* (33), 10664.
(86) Sun, N.; Liang, Y.; Ma, X.; Chen, F. Reduced Oxygenated g-C3N4 with Abundant Nitrogen Vacancies for Visible-Light Photocatalytic Applications. *Chemistry–A European Journal* **2017,** *23* (61), 15466.
(87) Fang, Y.; Li, X.; Wang, X. The synthesis of polymeric carbon nitride films with adhesive interfaces for solar water splitting devices. *ACS Catal.* **2018,** *8* (9), 8774.
(88) Wang, X.; Blechert, S.; Antonietti, M. Polymeric graphitic carbon nitride for heterogeneous photocatalysis. *ACS Catal.* **2012,** *2* (8), 1596.
(89) Miao, H.; Zhang, G.; Hu, X.; Mu, J.; Han, T.; Fan, J.; Zhu, C.; Song, L.; Bai, J.; Hou, X. A novel strategy to prepare 2D g-C3N4 nanosheets and their photoelectrochemical properties. *J. Alloys Compd.* **2017,** *690*, 669.
(90) Shalom, M.; Gimenez, S.; Schipper, F.; Herraiz-Cardona, I.; Bisquert, J.; Antonietti, M. Controlled carbon nitride growth on surfaces for hydrogen evolution electrodes. *Angew. Chem.* **2014,** *126* (14), 3728.
(91) Xu, J.; Brenner, T. J.; Chabanne, L.; Neher, D.; Antonietti, M.; Shalom, M. Liquid-based growth of polymeric carbon nitride layers and their use in a mesostructured polymer solar cell with V oc exceeding 1 V. *J. Am. Chem. Soc.* **2014,** *136* (39), 13486.
(92) Zhang, W.; Albero, J.; Xi, L.; Lange, K. M.; Garcia, H.; Wang, X.; Shalom, M. One-pot Synthesis of Nickel-Modified Carbon Nitride Layers Toward Efficient Photoelectrochemical Cells. *ACS Appl. Mater. Interfaces* **2017,** *9* (38), 32667.

(93) Ye, L.; Chen, S. Fabrication and high visible-light-driven photocurrent response of g-C3N4 film: The role of thiourea. *Appl. Surf. Sci.* **2016,** *389*, 1076.
(94) Xu, J.; Cao, S.; Brenner, T.; Yang, X.; Yu, J.; Antonietti, M.; Shalom, M. Supramolecular chemistry in molten sulfur: preorganization effects leading to marked enhancement of carbon nitride photoelectrochemistry. *Adv. Funct. Mater.* **2015,** *25* (39), 6265.
(95) Jun, Y. S.; Lee, E. Z.; Wang, X.; Hong, W. H.; Stucky, G. D.; Thomas, A. From melamine-cyanuric acid supramolecular aggregates to carbon nitride hollow spheres. *Adv. Funct. Mater.* **2013,** *23* (29), 3661.
(96) Bian, J.; Li, J.; Kalytchuk, S.; Wang, Y.; Li, Q.; Lau, T. C.; Niehaus, T. A.; Rogach, A. L.; Zhang, R. Q. Efficient emission facilitated by multiple energy level transitions in uniform graphitic carbon nitride films deposited by thermal vapor condensation. *ChemPhysChem* **2015,** *16* (5), 954.
(97) Bian, J.; Li, Q.; Huang, C.; Li, J.; Guo, Y.; Zaw, M.; Zhang, R.-Q. Thermal vapor condensation of uniform graphitic carbon nitride films with remarkable photocurrent density for photoelectrochemical applications. *Nano Energy* **2015,** *15*, 353.
(98) Bian, J.; Xi, L.; Huang, C.; Lange, K. M.; Zhang, R. Q.; Shalom, M. Efficiency Enhancement of Carbon Nitride Photoelectrochemical Cells via Tailored Monomers Design. *Adv. Energy Mater.* **2016,** *6* (12), 1600263.
(99) Lu, X.; Liu, Z.; Li, J.; Zhang, J.; Guo, Z. Novel framework g-C3N4 film as efficient photoanode for photoelectrochemical water splitting. *Appl. Catal. B* **2017,** *209*, 657.
(100) Ruan, Q.; Luo, W.; Xie, J.; Wang, Y.; Liu, X.; Bai, Z.; Carmalt, C. J.; Tang, J. A nanojunction polymer photoelectrode for efficient charge transport and separation. *Angew. Chem. Int. Ed.* **2017,** *56* (28), 8221.
(101) Lv, X.; Cao, M.; Shi, W.; Wang, M.; Shen, Y. A new strategy of preparing uniform graphitic carbon nitride films for photoelectrochemical application. *Carbon* **2017,** *117*, 343.
(102) Zada, A.; Humayun, M.; Raziq, F.; Zhang, X.; Qu, Y.; Bai, L.; Qin, C.; Jing, L.; Fu, H. Exceptional Visible-Light-Driven Cocatalyst-Free Photocatalytic Activity of g-C3N4 by Well Designed Nanocomposites with Plasmonic Au and SnO2. *Adv. Energy Mater.* **2016,** *6* (21), 1601190.
(103) Lim, E. L.; Yap, C. C.; Teridi, M. A. M.; Teh, C. H.; bin Mohd Yusoff, A. R.; Jumali, M. H. H. A review of recent plasmonic nanoparticles incorporated P3HT: PCBM organic thin film solar cells. *Org. Electron.* **2016,** *36*, 12.
(104) Teridi, M. A. M.; Sookhakian, M.; Basirun, W. J.; Zakaria, R.; Schneider, F. K.; Da Silva, W. J.; Kim, J.; Lee, S. J.; Kim, H. P.; bin Mohd Yusoff, A. R. Plasmon enhanced organic devices utilizing highly ordered nanoimprinted gold nanodisks and nitrogen doped graphene. *Nanoscale* **2015,** *7* (16), 7091.
(105) Xie, X.; Fan, X.; Huang, X.; Wang, T.; He, J. In situ growth of graphitic carbon nitride films on transparent conducting substrates via a solvothermal route for photoelectrochemical performance. *RSC Adv.* **2016,** *6* (12), 9916.
(106) Gu, Q.; Gong, X.; Jia, Q.; Liu, J.; Gao, Z.; Wang, X.; Long, J.; Xue, C. Compact carbon nitride based copolymer films with controllable thickness for photoelectrochemical water splitting. *J. Mater. Chem. A* **2017,** *5* (36), 19062.
(107) Xu, J.; Shalom, M. Electrophoretic deposition of carbon nitride layers for photoelectrochemical applications. *ACS Appl. Mater. Interfaces* **2016,** *8* (20), 13058.
(108) Dong, Y.; Chen, Y.; Jiang, P.; Wang, G.; Wu, X.; Wu, R. A novel gC 3 N 4 based photocathode for photoelectrochemical hydrogen evolution. *RSC Adv.* **2016,** *6* (9), 7465.
(109) Li, C.; Cao, C.-B.; Zhu, H.-S. Graphitic carbon nitride thin films deposited by electrodeposition. *Mater. Lett.* **2004,** *58* (12-13), 1903.
(110) Kang, D.; Kim, T. W.; Kubota, S. R.; Cardiel, A. C.; Cha, H. G.; Choi, K.-S. Electrochemical synthesis of photoelectrodes and catalysts for use in solar water splitting. *Chem. Rev.* **2015,** *115* (23), 12839.

(111) Hou, Y.; Wen, Z.; Cui, S.; Feng, X.; Chen, J. Strongly coupled ternary hybrid aerogels of N-deficient porous graphitic-C3N4 nanosheets/N-doped graphene/NiFe-layered double hydroxide for solar-driven photoelectrochemical water oxidation. *Nano Lett.* **2016,** *16* (4), 2268.
(112) Samanta, S.; Martha, S.; Parida, K. Facile Synthesis of Au/g-C3N4 Nanocomposites: An Inorganic/Organic Hybrid Plasmonic Photocatalyst with Enhanced Hydrogen Gas Evolution Under Visible-Light Irradiation. *ChemCatChem* **2014,** *6* (5), 1453.
(113) Di, Y.; Wang, X.; Thomas, A.; Antonietti, M. Making Metal-Carbon Nitride Heterojunctions for Improved Photocatalytic Hydrogen Evolution with Visible Light. *ChemCatChem* **2010,** *2* (7), 834.
(114) Liu, J.; Wang, H.; Chen, Z. P.; Moehwald, H.; Fiechter, S.; van de Krol, R.; Wen, L.; Jiang, L.; Antonietti, M. Microcontact-Printing-Assisted Access of Graphitic Carbon Nitride Films with Favorable Textures toward Photoelectrochemical Application. *Adv. Mater.* **2015,** *27* (4), 712.
(115) Chen, Z.; Wang, H.; Xu, J.; Liu, J. Surface Engineering of Carbon Nitride Electrode by Molecular Cobalt Species and Their Photoelectrochemical Application. *Chem. Asian J.* **2018,** *13* (12), 1539.
(116) Zhang, X.; Xie, X.; Wang, H.; Zhang, J.; Pan, B.; Xie, Y. Enhanced photoresponsive ultrathin graphitic-phase C3N4 nanosheets for bioimaging. *J. Am. Chem. Soc.* **2013,** *135* (1), 18.
(117) Wang, Y.; Zhao, X.; Tian, Y.; Wang, Y.; Jan, A. K.; Chen, Y. Facile Electrospinning Synthesis of Carbonized Polyvinylpyrrolidone (PVP)/g-C3N4 Hybrid Films for Photoelectrochemical Applications. *Chem. Eur. J.* **2017,** *23* (2), 419.
(118) Peng, G.; Volokh, M.; Tzadikov, J.; Sun, J.; Shalom, M. Carbon Nitride/Reduced Graphene Oxide Film with Enhanced Electron Diffusion Length: An Efficient Photo-Electrochemical Cell for Hydrogen Generation. *Adv. Energy Mater.* **2018**, 1800566.
(119) Wang, X. L.; Fang, W. Q.; Wang, H. F.; Zhang, H.; Zhao, H.; Yao, Y.; Yang, H. G. Surface hydrogen bonding can enhance photocatalytic H 2 evolution efficiency. *J. Mater. Chem. A* **2013,** *1* (45), 14089.
(120) Sano, T.; Tsutsui, S.; Koike, K.; Hirakawa, T.; Teramoto, Y.; Negishi, N.; Takeuchi, K. Activation of graphitic carbon nitride (gC 3 N 4) by alkaline hydrothermal treatment for photocatalytic NO oxidation in gas phase. *J. Mater. Chem. A* **2013,** *1* (21), 6489.
(121) Nie, H.; Ou, M.; Zhong, Q.; Zhang, S.; Yu, L. Efficient visible-light photocatalytic oxidation of gaseous NO with graphitic carbon nitride (g–C3N4) activated by the alkaline hydrothermal treatment and mechanism analysis. *J. Hazard. Mater.* **2015,** *300*, 598.
(122) Maleki, A.; Mohammad, M.; Emdadi, Z.; Asim, N.; Azizi, M.; Safaei, J. Adsorbent materials based on a geopolymer paste for dye removal from aqueous solutions. *Arabian J. Chem.* **2018. https://doi.org/10.1016/j.arabjc.2018.08.011**
(123) Nowotny, J.; Alim, M. A.; Bak, T.; Idris, M. A.; Ionescu, M.; Prince, K.; Sahdan, M. Z.; Sopian, K.; Teridi, M. A. M.; Sigmund, W. Defect chemistry and defect engineering of TiO 2-based semiconductors for solar energy conversion. *Chem. Soc. Rev.* **2015,** *44* (23), 8424.
(124) Algara-Siller, G.; Severin, N.; Chong, S. Y.; Björkman, T.; Palgrave, R. G.; Laybourn, A.; Antonietti, M.; Khimyak, Y. Z.; Krasheninnikov, A. V.; Rabe, J. P. Triazine-Based Graphitic Carbon Nitride: a Two-Dimensional Semiconductor. *Angew. Chem.* **2014,** *126* (29), 7580.
(125) Liu, N.; Han, M.; Sun, Y.; Zhu, C.; Zhou, Y.; Zhang, Y.; Huang, H.; Kremnican, V.; Liu, Y.; Lifshitz, Y. A gC 3 N 4 based photoelectrochemical cell using O 2/H 2 O redox couples. *Energy Environ. Sci.* **2018**.
(126) Liu, L.; Wang, M.; Wang, C. In-situ synthesis of graphitic carbon nitride/iron oxide-copper composites and their application in the electrochemical detection of glucose. *Electrochim. Acta* **2018,** *265*, 275.
(127) Zhao, F.; Cheng, H.; Hu, Y.; Song, L.; Zhang, Z.; Jiang, L.; Qu, L. Functionalized graphitic carbon nitride for metal-free, flexible and rewritable nonvolatile memory device via direct laser-writing. *Sci. Rep.* **2014,** *4*, 5882.

(128) Mohamad Noh, M. F.; Soh, M. F.; Riza, M. A.; Safaei, J.; Mohd Nasir, S. N.; Mohamad Sapian, N. W.; Teh, C. H.; Ibrahim, M. A.; Ahmad Ludin, N.; Mat Teridi, M. A. Effect of film thickness on photoelectrochemical performance of SnO2 prepared via AACVD. *Phys. Status Solidi B* **2018.** https://doi.org/10.1002/pssb.201700570.

(129) Zhan, F.; Xie, R.; Li, W.; Li, J.; Yang, Y.; Li, Y.; Chen, Q. In situ synthesis of gC 3 N 4/WO 3 heterojunction plates array films with enhanced photoelectrochemical performance. *RSC Adv.* **2015,** *5* (85), 69753.

(130) Su, J.; Geng, P.; Li, X.; Zhao, Q.; Quan, X.; Chen, G. Novel phosphorus doped carbon nitride modified TiO 2 nanotube arrays with improved photoelectrochemical performance. *Nanoscale* **2015,** *7* (39), 16282.

(131) Fang, Y.; Xu, Y.; Li, X.; Ma, Y.; Wang, X. Coating polymeric carbon nitride photoanodes on conductive Y: ZnO nanorod arrays for overall water splitting. *Angew. Chem.* **2018,** *130* (131), 9897.

(132) Feng, C.; Wang, Z.; Ma, Y.; Zhang, Y.; Wang, L.; Bi, Y. Ultrathin graphitic C3N4 nanosheets as highly efficient metal-free cocatalyst for water oxidation. *Appl. Catal. B* **2017,** *205*, 19.

(133) Basu, M.; Zhang, Z.-W.; Chen, C.-J.; Lu, T.-H.; Hu, S.-F.; Liu, R.-S. CoSe2 embedded in C3N4: an efficient photocathode for photoelectrochemical water splitting. *ACS Appl. Mater. Interfaces* **2016,** *8* (40), 26690.

(134) Xiao, J.; Zhang, X.; Li, Y. A ternary g-C3N4/Pt/ZnO photoanode for efficient photoelectrochemical water splitting. *Int. J. Hydrogen Energy* **2015,** *40* (30), 9080.

(135) Xu, X.; Liu, Y.; Zhu, Y.; Fan, X.; Li, Y.; Zhang, F.; Zhang, G.; Peng, W. Fabrication of a Cu2O/g-C3N4/WS2 Triple-Layer Photocathode for Photoelectrochemical Hydrogen Evolution. *ChemElectroChem* **2017,** *4* (6), 1498.

(136) Hajduk, Š.; Berglund, S. P.; Podlogar, M.; Dražić, G.; Abdi, F. F.; Orel, Z. C.; Shalom, M. Conformal Carbon Nitride Coating as an Efficient Hole Extraction Layer for ZnO Nanowires-Based Photoelectrochemical Cells. *Adv. Mater. Interfaces* **2017,** *4* (24), 1700924.

(137) Liu, Y.; Su, F.-Y.; Yu, Y.-X.; Zhang, W.-D. Nano g-C3N4 modified Ti-Fe2O3 vertically arrays for efficient photoelectrochemical generation of hydrogen under visible light. *Int. J. Hydrogen Energy* **2016,** *41* (18), 7270.

(138) Luo, J.; Dong, G.; Zhu, Y.; Yang, Z.; Wang, C. Switching of semiconducting behavior from n-type to p-type induced high photocatalytic NO removal activity in g-C3N4. *Appl. Catal. B* **2017,** *214*, 46.

(139) Basu, M.; Zhang, Z. W.; Chen, C. J.; Chen, P. T.; Yang, K. C.; Ma, C. G.; Lin, C. C.; Hu, S. F.; Liu, R. S. Heterostructure of Si and CoSe2: A Promising Photocathode Based on a Non-noble Metal Catalyst for Photoelectrochemical Hydrogen Evolution. *Angew. Chem. Int. Ed.* **2015,** *54* (21), 6211.

(140) Li, Y.; Wei, X.; Li, H.; Wang, R.; Feng, J.; Yun, H.; Zhou, A. Fabrication of inorganic–organic core–shell heterostructure: novel CdS@ gC 3 N 4 nanorod arrays for photoelectrochemical hydrogen evolution. *RSC Adv.* **2015,** *5* (19), 14074.

(141) Xu, J.; Herraiz-Cardona, I.; Yang, X.; Gimenez, S.; Antonietti, M.; Shalom, M. The complex role of carbon nitride as a sensitizer in photoelectrochemical cells. *Adv. Opt. Mater.* **2015,** *3* (8), 1052.

(142) Zhang, G.; Lan, Z.-A.; Wang, X. Surface engineering of graphitic carbon nitride polymers with cocatalysts for photocatalytic overall water splitting. *Chem. Sci.* **2017,** *8* (8), 5261.

(143) Liu, Y.; Yu, Y.-X.; Zhang, W.-D. Photoelectrochemical study on charge transfer properties of nanostructured Fe2O3 modified by g-C3N4. *Int. J. Hydrogen Energy* **2014,** *39* (17), 9105.

(144) Kuang, P.-Y.; Su, Y.-Z.; Chen, G.-F.; Luo, Z.; Xing, S.-Y.; Li, N.; Liu, Z.-Q. g-C3N4 decorated ZnO nanorod arrays for enhanced photoelectrocatalytic performance. *Appl. Surf. Sci.* **2015,** *358*, 296.

(145) Yang, F.; Lublow, M.; Orthmann, S.; Merschjann, C.; Tyborski, T.; Rusu, M.; Kubala, S.; Thomas, A.; Arrigo, R.; Hävecker, M. Metal-Free Photocatalytic Graphitic Carbon Nitride on

p-Type Chalcopyrite as a Composite Photocathode for Light-Induced Hydrogen Evolution. *ChemSusChem* **2012,** *5* (7), 1227.

(146) Yang, F.; Kuznietsov, V.; Lublow, M.; Merschjann, C.; Steigert, A.; Klaer, J.; Thomas, A.; Schedel-Niedrig, T. Solar hydrogen evolution using metal-free photocatalytic polymeric carbon nitride/CuInS 2 composites as photocathodes. *J. Mater. Chem. A* **2013,** *1* (21), 6407.

(147) Wang, J.; Zhang, W.-D. Modification of TiO2 nanorod arrays by graphite-like C3N4 with high visible light photoelectrochemical activity. *Electrochim. Acta* **2012,** *71*, 10.

(148) An, T.; Tang, J.; Zhang, Y.; Quan, Y.; Gong, X.; Al-Enizi, A. M.; Elzatahry, A. A.; Zhang, L.; Zheng, G. Photoelectrochemical Conversion from Graphitic C3N4 Quantum Dot Decorated Semiconductor Nanowires. *ACS Appl. Mater. Interfaces* **2016,** *8* (20), 12772.

(149) Sun, M.; Fang, Y.; Kong, Y.; Sun, S.; Yu, Z.; Umar, A. Graphitic carbon nitride (gC 3 N 4) coated titanium oxide nanotube arrays with enhanced photo-electrochemical performance. *Dalton Trans.* **2016,** *45* (32), 12702.

(150) Li, G.; Lian, Z.; Wang, W.; Zhang, D.; Li, H. Nanotube-confinement induced size-controllable g-C3N4 quantum dots modified single-crystalline TiO2 nanotube arrays for stable synergetic photoelectrocatalysis. *Nano Energy* **2016,** *19*, 446.

(151) Mustakim, N. S. M.; Ubani, C. A.; Sepeai, S.; Ludin, N. A.; Teridi, M. A. M.; Ibrahim, M. A. Quantum dots processed by SILAR for solar cell applications. *Sol. Energy* **2018,** *163*, 256.

(152) Kouhnavard, M.; Ikeda, S.; Ludin, N. A.; Khairudin, N. A.; Ghaffari, B.; Mat-Teridi, M.; Ibrahim, M. A.; Sepeai, S.; Sopian, K. A review of semiconductor materials as sensitizers for quantum dot-sensitized solar cells. *Renew. Sustain. Energy Rev.* **2014,** *37*, 397.

(153) Wang, W.; Jimmy, C. Y.; Shen, Z.; Chan, D. K.; Gu, T. gC 3 N 4 quantum dots: direct synthesis, upconversion properties and photocatalytic application. *Chem. Commun.* **2014,** *50* (70), 10148.

(154) Chen, Z.; Ma, G.; Chen, Z.; Zhang, Y.; Zhang, Z.; Gao, J.; Meng, Q.; Yuan, M.; Wang, X.; Liu, J.-m. Fabrication and photoelectrochemical properties of silicon nanowires/g-C3N4 core/shell arrays. *Appl. Surf. Sci.* **2017,** *396*, 609.

(155) Li, Y.; Wang, R.; Li, H.; Wei, X.; Feng, J.; Liu, K.; Dang, Y.; Zhou, A. Efficient and stable photoelectrochemical seawater splitting with TiO2@ g-C3N4 nanorod arrays decorated by Co-Pi. *J. Phys. Chem. C* **2015,** *119* (35), 20283.

(156) Li, H.; Zhao, F.; Zhang, J.; Luo, L.; Xiao, X.; Huang, Y.; Ji, H.; Tong, Y. A gC 3 N 4/WO 3 photoanode with exceptional ability for photoelectrochemical water splitting. *Mater. Chem. Front.* **2017,** *1* (2), 338.

(157) Hou, Y.; Zuo, F.; Dagg, A. P.; Liu, J.; Feng, P. Branched WO3 nanosheet array with layered C3N4 heterojunctions and CoOx nanoparticles as a flexible photoanode for efficient photoelectrochemical water oxidation. *Adv. Mater.* **2014,** *26* (29), 5043.

(158) Hou, J.; Cheng, H.; Takeda, O.; Zhu, H. Unique 3D heterojunction photoanode design to harness charge transfer for efficient and stable photoelectrochemical water splitting. *Energy Environ. Sci.* **2015,** *8* (4), 1348.

(159) Li, Y.; Wei, X.; Yan, X.; Cai, J.; Zhou, A.; Yang, M.; Liu, K. Construction of inorganic–organic 2D/2D WO 3/gC 3 N 4 nanosheet arrays toward efficient photoelectrochemical splitting of natural seawater. *Phys. Chem. Chem. Phys.* **2016,** *18* (15), 10255.

(160) Su, J.; Zhu, L.; Chen, G. Ultrasmall graphitic carbon nitride quantum dots decorated self-organized TiO2 nanotube arrays with highly efficient photoelectrochemical activity. *Appl. Catal. B* **2016,** *186*, 127.

(161) Wu, S.-Z.; Chen, C.-H.; Zhang, W.-D. Etching graphitic carbon nitride by acid for enhanced photocatalytic activity toward degradation of 4-nitrophenol. *Chin. Chem. Lett.* **2014,** *25* (9), 1247.

(162) Zhang, S.; Yan, J.; Yang, S.; Xu, Y.; Cai, X.; Li, X.; Zhang, X.; Peng, F.; Fang, Y. Electrodeposition of Cu2O/g-C3N4 heterojunction film on an FTO substrate for enhancing visible light photoelectrochemical water splitting. *Chin. J. Chem. Eng.* **2017,** *38* (2), 365.

(163) Wang, D.; Wang, C.; de Arquer, F. P. G.; Zhong, J.; Qian, L.; Fang, L.; Liu, P.; Pang, Y.; Liu, M.; Liu, M. Band-aligned C 3 N 4− x S 3x/2 stabilizes CdS/CuInGaS 2 photocathodes for efficient water reduction. *J. Mater. Chem. A* **2017,** *5* (7), 3167.
(164) Ye, L.; Wang, D.; Chen, S. Fabrication and enhanced photoelectrochemical performance of MoS2/S-doped g-C3N4 heterojunction film. *ACS Appl. Mater. Interfaces* **2016,** *8* (8), 5280.
(165) Zhou, P.; Yu, J.; Jaroniec, M. All-solid-state Z-scheme photocatalytic systems. *Adv. Mater.* **2014,** *26* (29), 4920.
(166) Dong, C.; Lu, S.; Yao, S.; Ge, R.; Wang, Z.; Wang, Z.; An, P.-F.; Liu, Y.; Yang, B.; Zhang, H. Colloidal Synthesis of Ultrathin Monoclinic BiVO4 Nanosheets for Z-Scheme Overall Water Splitting under Visible Light. *ACS Catal.* **2018**.
(167) Zeng, X.; Wang, Z.; Wang, G.; Gengenbach, T. R.; McCarthy, D. T.; Deletic, A.; Yu, J.; Zhang, X. Highly dispersed TiO2 nanocrystals and WO3 nanorods on reduced graphene oxide: Z-scheme photocatalysis system for accelerated photocatalytic water disinfection. *Appl. Catal. B* **2017,** *218*, 163.
(168) Wang, Y.; Suzuki, H.; Xie, J.; Tomita, O.; Martin, D. J.; Higashi, M.; Kong, D.; Abe, R.; Tang, J. Mimicking Natural Photosynthesis: Solar to Renewable H2 Fuel Synthesis by Z-Scheme Water Splitting Systems. *Chem. Rev.* **2018**.
(169) Yu, W.; Xu, D.; Peng, T. Enhanced photocatalytic activity of gC 3 N 4 for selective CO 2 reduction to CH 3 OH via facile coupling of ZnO: a direct Z-scheme mechanism. *J. Mater. Chem. A* **2015,** *3* (39), 19936.
(170) Wang, Q.; Hisatomi, T.; Jia, Q.; Tokudome, H.; Zhong, M.; Wang, C.; Pan, Z.; Takata, T.; Nakabayashi, M.; Shibata, N. Scalable water splitting on particulate photocatalyst sheets with a solar-to-hydrogen energy conversion efficiency exceeding 1%. *Nat. Mater.* **2016,** *15* (6), 611.
(171) Wang, Q.; Hisatomi, T.; Suzuki, Y.; Pan, Z.; Seo, J.; Katayama, M.; Minegishi, T.; Nishiyama, H.; Takata, T.; Seki, K. Particulate photocatalyst sheets based on carbon conductor layer for efficient Z-scheme pure-water splitting at ambient pressure. *J. Am. Chem. Soc.* **2017,** *139* (4), 1675.
(172) Wang, Q.; Hisatomi, T.; Katayama, M.; Takata, T.; Minegishi, T.; Kudo, A.; Yamada, T.; Domen, K. Particulate photocatalyst sheets for Z-scheme water splitting: advantages over powder suspension and photoelectrochemical systems and future challenges. *Faraday Discuss.* **2017,** *197*, 491.
(173) Park, J. H.; Yoon, J. S.; Oh, S. Y. Characterization of a Photoelectrochemical Cell Using Porous TiO2 and Its Application to a Z-Scheme System for Water Splitting in Combination with a P-Type InP Photocathode. *J. Nanosci. Nanotechnol.* **2017,** *17* (8), 5593.
(174) Arai, T.; Sato, S.; Kajino, T.; Morikawa, T. Solar CO 2 reduction using H2O by a semiconductor/metal-complex hybrid photocatalyst: enhanced efficiency and demonstration of a wireless system using SrTiO 3 photoanodes. *Energy Environ. Sci.* **2013,** *6* (4), 1274.
(175) Cho, K.-H.; Sung, Y.-M. The formation of Z-scheme CdS/CdO nanorods on FTO substrates: The shell thickness effects on the flat band potentials. *Nano Energy* **2017,** *36*, 176.
(176) Šutka, A.; Vanags, M.; Joost, U.; Šmits, K.; Ruža, J.; Ločs, J.; Kleperis, J.; Juhna, T. Aqueous synthesis of Z-scheme photocatalyst powders and thin-film photoanodes from earth abundant elements. *Journal of Environmental Chemical Engineering* **2018,** *6* (2), 2606.
(177) Cong, Y.; Ge, Y.; Zhang, T.; Wang, Q.; Shao, M.; Zhang, Y. Fabrication of Z-scheme Fe2O3-MoS2-Cu2O ternary nanofilm with significantly enhanced photoelectrocatalytic performance. *Ind. Eng. Chem. Res.* **2018**.
(178) Wang, Y.; Sun, J.; Li, J.; Zhao, X. Electrospinning Preparation of Nanostructured g-C3N4/BiVO4 Composite Films with an Enhanced Photoelectrochemical Performance. *Langmuir* **2017,** *33* (19), 4694.

(179) Huang, Z.; Zeng, X.; Li, K.; Gao, S.; Wang, Q.; Lu, J. Z-Scheme NiTiO3/g-C3N4 Heterojunctions with Enhanced Photoelectrochemical and Photocatalytic Performances under Visible LED Light Irradiation. *ACS Appl. Mater. Interfaces* **2017,** *9* (47), 41120.
(180) Zhang, B.; Zhao, S.-Y.; Wang, H.-H.; Zhao, T.-J.; Liu, Y.-X.; Lv, L.-B.; Wei, X.; Li, X.-H.; Chen, J.-S. The solution-phase process of a gC 3 N 4/BiVO 4 dyad to a large-area photoanode: interfacial synergy for highly efficient water oxidation. *Chem. Commun.* **2017,** *53* (76), 10544.
(181) Li, J.-M.; Cheng, H.-Y.; Chiu, Y.-H.; Hsu, Y.-J. ZnO–Au–SnO 2 Z-scheme photoanodes for remarkable photoelectrochemical water splitting. *Nanoscale* **2016,** *8* (34), 15720.
(182) Li, K.; Zeng, X.; Gao, S.; Ma, L.; Wang, Q.; Xu, H.; Wang, Z.; Huang, B.; Dai, Y.; Lu, J. Ultrasonic-assisted pyrolyzation fabrication of reduced SnO2–x/g-C3N4 heterojunctions: Enhance photoelectrochemical and photocatalytic activity under visible LED light irradiation. *Nano Res.* **2016,** *9* (7), 1969.
(183) Nasir, S. N. F. M.; Ullah, H.; Ebadi, M.; Tahir, A. A.; Sagu, J. S.; Mat Teridi, M. A. New Insights into Se/BiVO4 Heterostructure for Photoelectrochemical Water Splitting: A Combined Experimental and DFT Study. *J. Phys. Chem. C* **2017,** *121* (11), 6218.
(184) Pihosh, Y.; Turkevych, I.; Mawatari, K.; Uemura, J.; Kazoe, Y.; Kosar, S.; Makita, K.; Sugaya, T.; Matsui, T.; Fujita, D. Photocatalytic generation of hydrogen by core-shell WO3/BiVO4 nanorods with ultimate water splitting efficiency. *Sci. Rep.* **2015,** *5*, 11141.
(185) Ebadi, M.; Mat-Teridi, M.; Sulaiman, M.; Basirun, W.; Asim, N.; Ludin, N. A.; Ibrahim, M. A.; Sopian, K. Electrodeposited p-type Co 3 O 4 with high photoelectrochemical performance in aqueous medium. *RSC Adv.* **2015,** *5* (46), 36820.
(186) Charles, U.; Ibrahim, M.; Teridi, M. Electrodeposition of organic–inorganic tri-halide perovskites solar cell. *J. Power Sources* **2018,** *378*, 717.
(187) Noh, M. F. M.; Soh, M. F.; Teh, C. H.; Lim, E. L.; Yap, C. C.; Ibrahim, M. A.; Ludin, N. A.; Teridi, M. A. M. Effect of temperature on the properties of SnO 2 layer fabricated via AACVD and its application in photoelectrochemical cells and organic photovoltaic devices. *Sol. Energy* **2017,** *158*, 474.
(188) Nasir, S. N. F. M.; Yahya, M. K. N.; Sapian, N. W. M.; Ludin, N. A.; Ibrahim, M. A.; Sopian, K.; Teridi, M. A. M. Heterojunction Cr 2 O 3/CuO: Ni photocathodes for enhanced photoelectrochemical performance. *RSC Adv.* **2016,** *6* (62), 56885.
(189) Qiu, Y.; Liu, W.; Chen, W.; Zhou, G.; Hsu, P.-C.; Zhang, R.; Liang, Z.; Fan, S.; Zhang, Y.; Cui, Y. Efficient solar-driven water splitting by nanocone BiVO 4-perovskite tandem cells. *Sci. Adv.* **2016,** *2* (6), e1501764.
(190) Zeng, D.; Wu, P.; Ong, W.-J.; Tang, B.; Wu, M.; Zheng, H.; Chen, Y.; Peng, D.-L. Construction of network-like and flower-like 2H-MoSe2 nanostructures coupled with porous g-C3N4 for noble-metal-free photocatalytic H2 evolution under visible light. *Appl. Catal. B* **2018,** *233*, 26.
(191) Jing, L.; Ong, W.-J.; Zhang, R.; Pickwell-MacPherson, E.; Jimmy, C. Y. Graphitic carbon nitride nanosheet wrapped mesoporous titanium dioxide for enhanced photoelectrocatalytic water splitting. *Catal. Today* **2018**.
(192) Ye, M. Y.; Zhao, Z. H.; Hu, Z. F.; Liu, L. Q.; Ji, H. M.; Shen, Z. R.; Ma, T. Y. 0D/2D Heterojunctions of Vanadate Quantum Dots/Graphitic Carbon Nitride Nanosheets for Enhanced Visible-Light-Driven Photocatalysis. *Angew. Chem. Int. Ed.* **2017,** *56* (29), 8407.
(193) Ansari, S. A.; Cho, M. H. Simple and Large Scale Construction of MoS2-g-C3N4 Heterostructures Using Mechanochemistry for High Performance Electrochemical Supercapacitor and Visible Light Photocatalytic Applications. *Sci. Rep.* **2017,** *7*, 43055.
(194) Bhandary, N.; Singh, A. P.; Ingole, P. P.; Basu, S. Enhancing the Photoelectrochemical Performance of a Hematite Dendrite/Graphitic Carbon Nitride Nanocomposite through Surface Modification with CoFeOx. *ChemPhotoChem* **2017,** *1* (2), 70.

(195) Arif, M.; Yasin, G.; Shakeel, M.; Fang, X.; Gao, R.; Ji, S.; Yan, D. Coupling of Bifunctional CoMn-Layered Double Hydroxide@ Graphitic C3N4 Nanohybrids towards Efficient Photoelectrochemical Overall Water Splitting. *Chem. Asian J.* **2018,** *13* (8), 1045.
(196) Han, M.; Zhu, S.; Lu, S.; Song, Y.; Feng, T.; Tao, S.; Liu, J.; Yang, B. Recent progress on the photocatalysis of carbon dots: Classification, mechanism and applications. *Nano Today* **2018,** *19*, 201.
(197) Liu, Y.; Zhang, H.; Ke, J.; Zhang, J.; Tian, W.; Xu, X.; Duan, X.; Sun, H.; Tade, M. O.; Wang, S. 0D (MoS2)/2D (g-C3N4) heterojunctions in Z-scheme for enhanced photocatalytic and electrochemical hydrogen evolution. *Appl. Catal. B* **2018,** *228*, 64.
(198) Kumar, A.; Schuerings, C.; Kumar, S.; Kumar, A.; Krishnan, V. Perovskite-structured CaTiO3 coupled with g-C3N4 as a heterojunction photocatalyst for organic pollutant degradation. *Beilstein journal of nanotechnology* **2018,** *9*, 671.
(199) Zhang, G.; Liu, G.; Wang, L.; Irvine, J. T. Inorganic perovskite photocatalysts for solar energy utilization. *Chem. Soc. Rev.* **2016,** *45* (21), 5951.
(200) Lou, S.; Zhou, Z.; Shen, Y.; Zhan, Z.; Wang, J.; Liu, S.; Zhang, Y. Comparison Study of the Photoelectrochemical Activity of Carbon Nitride with Different Photoelectrode Configurations. *ACS Appl. Mater. Interfaces* **2016,** *8* (34), 22287.
(201) Xiao, M.; Wang, Z.; Lyu, M.; Luo, B.; Wang, S.; Liu, G.; Cheng, H. M.; Wang, L. Hollow Nanostructures for Photocatalysis: Advantages and Challenges. *Adv. Mater.* **2018**, 1801369.
(202) Sun, J.; Zhang, J.; Zhang, M.; Antonietti, M.; Fu, X.; Wang, X. Bioinspired hollow semiconductor nanospheres as photosynthetic nanoparticles. *Nat. Comm.* **2012,** *3*, 1139.
(203) Cui, Y.; Ding, Z.; Fu, X.; Wang, X. Construction of conjugated carbon nitride nanoarchitectures in solution at low temperatures for photoredox catalysis. *Angew. Chem.* **2012,** *124* (47), 11984.
(204) Zeng, D.; Ong, W. J.; Chen, Y.; Tee, S. Y.; Chua, C. S.; Peng, D. L.; Han, M. Y. Co2P Nanorods as an Efficient Cocatalyst Decorated Porous g-C3N4 Nanosheets for Photocatalytic Hydrogen Production under Visible Light Irradiation. *Part. Part. Syst. Charact.* **2018,** *35* (1), 1700251.
(205) Wang, J.; Tang, L.; Zeng, G.; Deng, Y.; Liu, Y.; Wang, L.; Zhou, Y.; Guo, Z.; Wang, J.; Zhang, C. Atomic scale g-C3N4/Bi2WO6 2D/2D heterojunction with enhanced photocatalytic degradation of ibuprofen under visible light irradiation. *Appl. Catal. B* **2017,** *209*, 285.
(206) Ong, W.-J. 2D/2D graphitic carbon nitride (g-C3N4) heterojunction nanocomposites for photocatalysis: why does face-to-face interface matter? *Frontiers in Materials* **2017,** *4*, 11.
(207) Wang, X.; Chen, X.; Thomas, A.; Fu, X.; Antonietti, M. Metal-Containing Carbon Nitride Compounds: A New Functional Organic–Metal Hybrid Material. *Adv. Mater.* **2009,** *21* (16), 1609.
(208) Bhandary, N.; Singh, A. P.; Kumar, S.; Ingole, P. P.; Thakur, G. S.; Ganguli, A. K.; Basu, S. In Situ Solid-State Synthesis of a AgNi/g-C3N4 Nanocomposite for Enhanced Photoelectrochemical and Photocatalytic Activity. *ChemSusChem* **2016,** *9* (19), 2816.
(209) Chen, X.; Zhang, J.; Fu, X.; Antonietti, M.; Wang, X. Fe-g-C3N4-catalyzed oxidation of benzene to phenol using hydrogen peroxide and visible light. *J. Am. Chem. Soc.* **2009,** *131* (33), 11658.
(210) Sagara, N.; Kamimura, S.; Tsubota, T.; Ohno, T. Photoelectrochemical CO2 reduction by a p-type boron-doped g-C3N4 electrode under visible light. *Appl. Catal. B* **2016,** *192*, 193.
(211) Lin, Z.; Wang, X. Nanostructure engineering and doping of conjugated carbon nitride semiconductors for hydrogen photosynthesis. *Angew. Chem.* **2013,** *125* (6), 1779.
(212) Zhang, Y.; Mori, T.; Ye, J.; Antonietti, M. Phosphorus-doped carbon nitride solid: enhanced electrical conductivity and photocurrent generation. *J. Am. Chem. Soc.* **2010,** *132* (18), 6294.
(213) Zhang, G.; Zhang, M.; Ye, X.; Qiu, X.; Lin, S.; Wang, X. Iodine modified carbon nitride semiconductors as visible light photocatalysts for hydrogen evolution. *Adv. Mater.* **2014,** *26* (5), 805.
(214) Zhang, Y.; Schnepp, Z.; Cao, J.; Ouyang, S.; Li, Y.; Ye, J.; Liu, S. Biopolymer-activated graphitic carbon nitride towards a sustainable photocathode material. *Sci. Rep.* **2013,** *3*, 2163.

(215) Jiang, L.; Yuan, X.; Pan, Y.; Liang, J.; Zeng, G.; Wu, Z.; Wang, H. Doping of graphitic carbon nitride for photocatalysis: a reveiw. *Appl. Catal. B* **2017,** *217*, 388.
(216) Ou, H.; Chen, X.; Lin, L.; Fang, Y.; Wang, X. Biomimetic Donor-Acceptor Motifs in Conjugated Polymers for Promoting Exciton Splitting and Charge Separation. *Angew. Chem. Int. Ed.* **2018.** https://doi.org/10.1002/anie.201803863.
(217) Zhou, L.; Xu, Y.; Yu, W.; Guo, X.; Yu, S.; Zhang, J.; Li, C. Ultrathin two-dimensional graphitic carbon nitride as a solution-processed cathode interfacial layer for inverted polymer solar cells. *J. Mater. Chem. A* **2016,** *4* (21), 8000.
(218) Chen, X.; Liu, Q.; Wu, Q.; Du, P.; Zhu, J.; Dai, S.; Yang, S. Incorporating Graphitic Carbon Nitride (g-C3N4) Quantum Dots into Bulk-Heterojunction Polymer Solar Cells Leads to Efficiency Enhancement. *Adv. Funct. Mater.* **2016,** *26* (11), 1719.
(219) Chetia, T. R.; Ansari, M. S.; Qureshi, M. Graphitic carbon nitride as a photovoltaic booster in quantum dot sensitized solar cells: a synergistic approach for enhanced charge separation and injection. *J. Mater. Chem. A* **2016,** *4* (15), 5528.
(220) Gao, Q.; Sun, S.; Li, X.; Zhang, X.; Duan, L.; Lü, W. Enhancing performance of CdS quantum dot-sensitized solar cells by two-dimensional gC 3 N 4 modified TiO 2 nanorods. *Nanoscale Res. Lett.* **2016,** *11* (1), 463.
(221) Xu, J.; Wang, G.; Fan, J.; Liu, B.; Cao, S.; Yu, J. g-C3N4 modified TiO2 nanosheets with enhanced photoelectric conversion efficiency in dye-sensitized solar cells. *J. Power Sources* **2015,** *274*, 77.
(222) Yan, H.; Tian, X.; Pang, Y.; Feng, B.; Duan, K.; Zhou, Z.; Weng, J.; Wang, J. Heterostructured gC 3 N 4/Ag/TiO 2 nanocomposites for enhancing the photoelectric conversion efficiency of spiro-OMeTAD-based solid-state dye-sensitized solar cells. *RSC Adv.* **2016,** *6* (104), 102444.
(223) Fan, K.; Yu, J.; Ho, W. Improving photoanodes to obtain highly efficient dye-sensitized solar cells: a brief review. *Materials Horizons* **2017,** *4* (3), 319.
(224) Ubani, C.; Ibrahim, M.; Teridi, M. Moving into the domain of perovskite sensitized solar cell. *Renew. Sustain. Energy Rev.* **2017,** *72*, 907.
(225) Yusoff, A. R. b. M.; Nazeeruddin, M. K. Organohalide lead perovskites for photovoltaic applications. *J. Phys. Chem. Lett* **2016,** *7* (5), 851.
(226) Hsieh, C.-M.; Liao, Y.-S.; Lin, Y.-R.; Chen, C.-P.; Tsai, C.-M.; Diau, E. W.-G.; Chuang, S.-C. Low-temperature, simple and efficient preparation of perovskite solar cells using Lewis bases urea and thiourea as additives: stimulating large grain growth and providing a PCE up to 18.8%. *RSC Adv.* **2018,** *8* (35), 19610.
(227) Jiang, L. L.; Wang, Z. K.; Li, M.; Zhang, C. C.; Ye, Q. Q.; Hu, K. H.; Lu, D. Z.; Fang, P. F.; Liao, L. S. Passivated Perovskite Crystallization via g-C3N4 for High-Performance Solar Cells. *Adv. Funct. Mater.* **2018,** *28* (7), 1705875.
(228) Noh, M. F. M.; Teh, C. H.; Daik, R.; Lim, E. L.; Yap, C. C.; Ibrahim, M. A.; Ludin, N. A.; bin Mohd Yusoff, A. R.; Jang, J.; Teridi, M. A. M. The architecture of the electron transport layer for a perovskite solar cell. *J. Mater. Chem. C* **2018,** *6*, 682.
(229) bin Mohd Yusoff, A. R.; Teridi, M. A. M.; Jang, J. Null current hysteresis for acetylacetonate electron extraction layer in perovskite solar cells. *Nanoscale* **2016,** *8* (12), 6328.
(230) Mohamad Noh, M. F.; Arzaee, N. A.; Safaei, J.; Mohamed, N. A.; Kim, H. P.; Yusoff, A. R. M.; Jang, J.; Teridi, M. Eliminating oxygen vacancies in SnO2 films via aerosol-assisted chemical vapour deposition for perovskite solar cells and photoelectrochemical cells. *J. Alloys Compd.* **2018.** https://doi.org/10.1016/j.jallcom.2018.09.273.
(231) Teh, C. H.; Daik, R.; Lim, E. L.; Yap, C. C.; Ibrahim, M. A.; Ludin, N. A.; Sopian, K.; Teridi, M. A. M. A review of organic small molecule-based hole-transporting materials for meso-structured organic–inorganic perovskite solar cells. *J. Mater. Chem. A* **2016,** *4* (41), 15788.
(232) Kim, J.; Teridi, M. A. M.; bin Mohd Yusoff, A. R.; Jang, J. Stable and null current hysteresis perovskite solar cells based nitrogen doped graphene oxide nanoribbons hole transport layer. *Sci. Rep.* **2016,** *6*, 27773.

(233) Zhang, K.; Ma, M.; Li, P.; Wang, D. H.; Park, J. H. Water splitting progress in tandem devices: moving photolysis beyond electrolysis. *Adv. Energy Mater.* **2016,** *6* (15), 1600602.
(234) Liang, J.; Zheng, Y.; Chen, J.; Liu, J.; Hulicova-Jurcakova, D.; Jaroniec, M.; Qiao, S. Z. Facile Oxygen Reduction on a Three-Dimensionally Ordered Macroporous Graphitic C3N4/Carbon Composite Electrocatalyst. *Angew. Chem.* **2012,** *124* (16), 3958.
(235) Duan, J.; Chen, S.; Jaroniec, M.; Qiao, S. Z. Porous C3N4 nanolayers@ N-graphene films as catalyst electrodes for highly efficient hydrogen evolution. *ACS Nano* **2015,** *9* (1), 931.
(236) Zou, X.; Silva, R.; Goswami, A.; Asefa, T. Cu-doped carbon nitride: Bio-inspired synthesis of H 2-evolving electrocatalysts using graphitic carbon nitride (gC 3 N 4) as a host material. *Appl. Surf. Sci.* **2015,** *357*, 221.
(237) Zheng, Y.; Jiao, Y.; Zhu, Y.; Li, L. H.; Han, Y.; Chen, Y.; Du, A.; Jaroniec, M.; Qiao, S. Z. Hydrogen evolution by a metal-free electrocatalyst. *Nat. Comm.* **2014,** *5*, 3783.
(238) Pan, L. F.; Li, Y. H.; Yang, S.; Liu, P. F.; Yu, M. Q.; Yang, H. G. Molybdenum carbide stabilized on graphene with high electrocatalytic activity for hydrogen evolution reaction. *Chem. Commun.* **2014,** *50* (86), 13135.
(239) Kouhnavard, M.; Ludin, N. A.; Ghaffari, B. V.; Sopian, K.; Ikeda, S. Carbonaceous Materials and Their Advances as a Counter Electrode in Dye-Sensitized Solar Cells: Challenges and Prospects. *ChemSusChem* **2015,** *8* (9), 1510.
(240) Wang, G.; Zhang, J.; Hou, S. g-C3N4/conductive carbon black composite as Pt-free counter electrode in dye-sensitized solar cells. *Mater. Res. Bull.* **2016,** *76*, 454.
(241) Wang, G.; Zhang, J.; Kuang, S.; Zhang, W. Enhanced Electrocatalytic Performance of a Porous g-C3N4/Graphene Composite as a Counter Electrode for Dye-Sensitized Solar Cells. *Chem. Eur. J.* **2016,** *22* (33), 11763.
(242) Afshari, M.; Dinari, M.; Momeni, M. M. Ultrasonic irradiation preparation of graphitic-C3N4/polyaniline nanocomposites as counter electrodes for dye-sensitized solar cells. *Ultrason. Sonochem.* **2018,** *42*, 631.
(243) Yuan, H.; Liu, J.; Li, H.; Li, Y.; Liu, X.; Shi, D.; Wu, Q.; Jiao, Q. Graphitic carbon nitride quantum dot decorated three-dimensional graphene as an efficient metal-free electrocatalyst for triiodide reduction. *J. Mater. Chem. A* **2018,** *6* (14), 5603.
(244) Tahir, M.; Cao, C.; Mahmood, N.; Butt, F. K.; Mahmood, A.; Idrees, F.; Hussain, S.; Tanveer, M.; Ali, Z.; Aslam, I. Multifunctional g-C3N4 nanofibers: a template-free fabrication and enhanced optical, electrochemical, and photocatalyst properties. *ACS Appl. Mater. Interfaces* **2013,** *6* (2), 1258.
(245) Tahir, M.; Cao, C.; Butt, F. K.; Idrees, F.; Mahmood, N.; Ali, Z.; Aslam, I.; Tanveer, M.; Rizwan, M.; Mahmood, T. Tubular graphitic-C 3 N 4: a prospective material for energy storage and green photocatalysis. *J. Mater. Chem. A* **2013,** *1* (44), 13949.
(246) Liang, Q.; Ye, L.; Xu, Q.; Huang, Z.-H.; Kang, F.; Yang, Q.-H. Graphitic carbon nitride nanosheet-assisted preparation of N-enriched mesoporous carbon nanofibers with improved capacitive performance. *Carbon* **2015,** *94*, 342.
(247) Shi, L.; Zhang, J.; Liu, H.; Que, M.; Cai, X.; Tan, S.; Huang, L. Flower-like Ni (OH) 2 hybridized g-C3N4 for high-performance supercapacitor electrode material. *Mater. Lett.* **2015,** *145*, 150.
(248) Zhang, L.; Ou, M.; Yao, H.; Li, Z.; Qu, D.; Liu, F.; Wang, J.; Wang, J.; Li, Z. Enhanced supercapacitive performance of graphite-like C3N4 assembled with NiAl-layered double hydroxide. *Electrochim. Acta* **2015,** *186*, 292.
(249) Dong, B.; Li, M.; Chen, S.; Ding, D.; Wei, W.; Gao, G.; Ding, S. Formation of g-C3N4@ Ni (OH) 2 honeycomb nanostructure and asymmetric supercapacitor with high energy and power density. *ACS Appl. Mater. Interfaces* **2017,** *9* (21), 17890.
(250) Adhikari, S. P.; Awasthi, G. P.; Kim, K.-S.; Park, C. H.; Kim, C. S. Synthesis of three-dimensional mesoporous Cu–Al layered double hydroxide/gC 3 N 4 nanocomposites on Ni-foam for enhanced supercapacitors with excellent long-term cycling stability. *Dalton Trans.* **2018,** *47* (13), 4455.

(251) Ding, Y.; Tang, Y.; Yang, L.; Zeng, Y.; Yuan, J.; Liu, T.; Zhang, S.; Liu, C.; Luo, S. Porous nitrogen-rich carbon materials from carbon self-repairing gC 3 N 4 assembled with graphene for high-performance supercapacitor. *J. Mater. Chem. A* **2016,** *4* (37), 14307.
(252) Lin, R.; Li, Z.; El Amaiem, D. I. A.; Zhang, B.; Brett, D. J.; He, G.; Parkin, I. P. A general method for boosting the supercapacitor performance of graphitic carbon nitride/graphene hybrids. *J. Mater. Chem. A* **2017,** *5* (48), 25545.
(253) Zhang, J.; Ding, J.; Li, C.; Li, B.; Li, D.; Liu, Z.; Cai, Q.; Zhang, J.; Liu, Y. Fabrication of novel ternary three-dimensional RuO2/graphitic-C3N4@ reduced graphene oxide aerogel composites for supercapacitors. *ACS Sustain. Chem. Eng.* **2017,** *5* (6), 4982.
(254) Xu, L.; Xia, J.; Xu, H.; Yin, S.; Wang, K.; Huang, L.; Wang, L.; Li, H. Reactable ionic liquid assisted solvothermal synthesis of graphite-like C3N4 hybridized α-Fe2O3 hollow microspheres with enhanced supercapacitive performance. *J. Power Sources* **2014,** *245*, 866.
(255) Chang, X.; Zhai, X.; Sun, S.; Gu, D.; Dong, L.; Yin, Y.; Zhu, Y. MnO2/g-C3N4 nanocomposite with highly enhanced supercapacitor performance. *Nanotechnology* **2017,** *28* (13), 135705.
(256) Zhao, Y.; Xu, L.; Huang, S.; Bao, J.; Qiu, J.; Lian, J.; Xu, L.; Huang, Y.; Xu, Y.; Li, H. Facile preparation of TiO2/C3N4 hybrid materials with enhanced capacitive properties for high performance supercapacitors. *J. Alloys Compd.* **2017,** *702*, 178.
(257) Wu, Y.; Chen, M.; Yan, X.; Ren, J.; Dai, Y.; Wang, J.; Pan, J.; Wang, Y.; Cheng, X. Hydrothermal synthesis of Fe3O4 nanorods/graphitic C3N4 composite with enhanced supercapacitive performance. *Mater. Lett.* **2017,** *198*, 114.
(258) Vattikuti, S. P.; Reddy, B. P.; Byon, C.; Shim, J. Carbon/CuO nanosphere-anchored g-C3N4 nanosheets as ternary electrode material for supercapacitors. *J. Solid State Chem.* **2018,** *262*, 106.
(259) Zhang, N.; Chen, C.; Chen, Y.; Chen, G.; Liao, C.; Liang, B.; Zhang, J.; Li, A.; Yang, B.; Zheng, Z. Ni2P2O7 Nanoarrays with Decorated C3N4 Nanosheets as Efficient Electrode for Supercapacitors. *ACS Appl. Energy Mater.* **2018,** *1* (5), 2016.
(260) Guo, W.; Wang, J.; Fan, C.; Chen, Z.; Liu, P.; Zhu, D.; Xu, Z.; Pang, L.; Li, T. Synthesis of Carbon Self-Repairing Porous g-C3N4 Nanosheets/NiCo2S4 Nanoparticles Hybrid Composite as High-Performance Electrode Materials for Supercapacitors. *Electrochim. Acta* **2017,** *253*, 68.
(261) Jiang, D.; Xu, Q.; Meng, S.; Xia, C.; Chen, M. Construction of cobalt sulfide/graphitic carbon nitride hybrid nanosheet composites for high performance supercapacitor electrodes. *J. Alloys Compd.* **2017,** *706*, 41.
(262) Ansari, S. A.; Cho, M. H. Growth of three-dimensional flower-like SnS 2 on gC 3 N 4 sheets as an efficient visible-light photocatalyst, photoelectrode, and electrochemical supercapacitance material. *Sustain. Energy Fuels* **2017,** *1* (3), 510.
(263) Wei, B.; Liang, H.; Wang, R.; Zhang, D.; Qi, Z.; Wang, Z. One-step synthesis of graphitic-C3N4/ZnS composites for enhanced supercapacitor performance. *J. Energy Chem.* **2018,** *27* (2), 472.
(264) Chen, X.; Zhu, X.; Xiao, Y.; Yang, X. PEDOT/g-C3N4 binary electrode material for supercapacitors. *J. Electroanal. Chem.* **2015,** *743*, 99.
(265) Li, Q.; Xu, D.; Guo, J.; Ou, X.; Yan, F. Protonated g-C3N4@ polypyrrole derived N-doped porous carbon for supercapacitors and oxygen electrocatalysis. *Carbon* **2017,** *124*, 599.
(266) Zhou, S.-X.; Tao, X.-Y.; Ma, J.; Guo, L.-T.; Zhu, Y.-B.; Fan, H.-L.; Liu, Z.-S.; Wei, X.-Y. Synthesis of flower-like PANI/g-C3N4 nanocomposite as supercapacitor electrode. *Vacuum* **2018,** *149*, 175.
(267) Shayeh, J. S.; Salari, H.; Daliri, A.; Omidi, M. Decorative reduced graphene oxide/C3N4/Ag2O/conductive polymer as a high performance material for electrochemical capacitors. *Appl. Surf. Sci.* **2018,** *447*, 374.
(268) Xie, R.-L.; Zong, Z.-M.; Liu, F.-J.; Wang, Y.-G.; Yan, H.-L.; Wei, Z.-H.; Mayyas, M.; Wei, X.-Y. Nitrogen-doped porous carbon foams prepared from mesophase pitch through graphitic carbon nitride nanosheet templates. *RSC Adv.* **2015,** *5* (57), 45718.

(269) Ansari, S. A.; Ansari, M. O.; Cho, M. H. Facile and scale up synthesis of red phosphorus-graphitic carbon nitride heterostructures for energy and environment applications. *Sci. Rep.* **2016,** *6*, 27713.
(270) Yoon, Y.; Lee, M.; Kim, S. K.; Bae, G.; Song, W.; Myung, S.; Lim, J.; Lee, S. S.; Zyung, T.; An, K. S. A Strategy for Synthesis of Carbon Nitride Induced Chemically Doped 2D MXene for High-Performance Supercapacitor Electrodes. *Adv. Energy Mater.* **2018,** *8* (15), 1703173.
(271) Barrio, J.; Lin, L.; Amo-Ochoa, P.; Tzadikov, J.; Peng, G.; Sun, J.; Zamora, F.; Wang, X.; Shalom, M. Unprecedented Centimeter-Long Carbon Nitride Needles: Synthesis, Characterization and Applications. *Small* **2018,** *14* (21), 1800633.
(272) Liu, Y.-H.; Xu, J.-L.; Gao, X.; Sun, Y.-L.; Lv, J.-J.; Shen, S.; Chen, L.-S.; Wang, S.-D. Freestanding transparent metallic network based ultrathin, foldable and designable supercapacitors. *Energy Environ. Sci.* **2017,** *10* (12), 2534.
(273) Zheng, Y.; Jiao, Y.; Zhu, Y.; Cai, Q.; Vasileff, A.; Li, L. H.; Han, Y.; Chen, Y.; Qiao, S.-Z. Molecule-level g-C3N4 coordinated transition metals as a new class of electrocatalysts for oxygen electrode reactions. *J. Am. Chem. Soc.* **2017,** *139* (9), 3336.
(274) Luo, W. B.; Chou, S. L.; Wang, J. Z.; Zhai, Y. C.; Liu, H. K. A Metal-Free, Free-Standing, Macroporous Graphene@ g-C3N4 Composite Air Electrode for High-Energy Lithium Oxygen Batteries. *Small* **2015,** *11* (23), 2817.
(275) Zheng, Y.; Jiao, Y.; Chen, J.; Liu, J.; Liang, J.; Du, A.; Zhang, W.; Zhu, Z.; Smith, S. C.; Jaroniec, M. Nanoporous graphitic-C3N4@ carbon metal-free electrocatalysts for highly efficient oxygen reduction. *J. Am. Chem. Soc.* **2011,** *133* (50), 20116.
(276) Liu, Q.; Zhang, J. Graphene supported Co-g-C3N4 as a novel metal–macrocyclic electrocatalyst for the oxygen reduction reaction in fuel cells. *Langmuir* **2013,** *29* (11), 3821.
(277) He, P.; Tang, X.; Chen, L.; Xie, P.; He, L.; Zhou, H.; Zhang, D.; Fan, T. Patterned Carbon Nitride–Based Hybrid Aerogel Membranes via 3D Printing for Broadband Solar Wastewater Remediation. *Adv. Funct. Mater.* **2018,** *28* (29), 1801121.
(278) Ou, H.; Yang, P.; Lin, L.; Anpo, M.; Wang, X. Carbon nitride aerogels for the photoredox conversion of water. *Angew. Chem.* **2017,** *129* (36), 11045.
(279) Xu, J.; Shalom, M.; Piersimoni, F.; Antonietti, M.; Neher, D.; Brenner, T. J. Color-Tunable Photoluminescence and NIR Electroluminescence in Carbon Nitride Thin Films and Light-Emitting Diodes. *Adv. Opt. Mater.* **2015,** *3* (7), 913.
(280) Gan, Z.; Shan, Y.; Chen, J.; Gui, Q.; Zhang, Q.; Nie, S.; Wu, X. The origins of the broadband photoluminescence from carbon nitrides and applications to white light emitting. *Nano Res.* **2016,** *9* (6), 1801.
(281) Zhang, X.; Zheng, Q.; Tang, Z.; Li, W.; Zhang, Y.; Xu, K.; Xue, X.; Xu, J.; Wang, H.; Wei, B. Tunable hole injection of solution-processed polymeric carbon nitride towards efficient organic light-emitting diode. *Appl. Phys. Lett.* **2018,** *112* (8), 083302.
(282) Velusamy, D. B.; Haque, M. A.; Parida, M. R.; Zhang, F.; Wu, T.; Mohammed, O. F.; Alshareef, H. N. 2D Organic–Inorganic Hybrid Thin Films for Flexible UV–Visible Photodetectors. *Adv. Funct. Mater.* **2017,** *27* (15), 1605554.
(283) Prakash, N.; Kumar, G.; Singh, M.; Barvat, A.; Pal, P.; Singh, S. P.; Singh, H.; Khanna, S. P. Binary Multifunctional Ultrabroadband Self-Powered g-C3N4/Si Heterojunction High-Performance Photodetector. *Adv. Opt. Mater.* **2018,** *6* (14), 1800191.
(284) Lai, S. K.; Xie, C.; Teng, K. S.; Li, Y.; Tan, F.; Yan, F.; Lau, S. P. Polymeric Carbon Nitride Nanosheets/Graphene Hybrid Phototransistors with High Responsivity. *Adv. Opt. Mater.* **2016,** *4* (4), 555.
(285) Zhuang, J.; Lai, W.; Xu, M.; Zhou, Q.; Tang, D. Plasmonic AuNP/g-C3N4 nanohybrid-based photoelectrochemical sensing platform for ultrasensitive monitoring of polynucleotide kinase activity accompanying DNAzyme-catalyzed precipitation amplification. *ACS Appl. Mater. Interfaces* **2015,** *7* (15), 8330.

(286) Dong, Y.-X.; Cao, J.; Wang, B.; Ma, S.-H.; Liu, Y. M. Spatial-Resolved Photoelectrochemical Biosensing Array Based on CdS@ g-C3N4 Heterojunction: A Universal Immunosensing Platform for Accurate Detection. *ACS Appl. Mater. Interfaces* **2018**.
(287) Liu, Y.; Yan, K.; Zhang, J. Graphitic carbon nitride sensitized with CdS quantum dots for visible-light-driven photoelectrochemical aptasensing of tetracycline. *ACS Appl. Mater. Interfaces* **2015,** *8* (42), 28255.
(288) Liu, Y.; Ma, H.; Zhang, Y.; Pang, X.; Fan, D.; Wu, D.; Wei, Q. Visible light photoelectrochemical aptasensor for adenosine detection based on CdS/PPy/g-C3N4 nanocomposites. *Biosens. Bioelectron.* **2016,** *86*, 439.
(289) Zeng, B.; Zhang, L.; Wan, X.; Song, H.; Lv, Y. Fabrication of α-Fe2O3/g-C3N4 composites for cataluminescence sensing of H2S. *Sens. Actuators B Chem.* **2015,** *211*, 370.
(290) Yan, J.; Rodrigues, M. T. F.; Song, Z.; Li, H.; Xu, H.; Liu, H.; Wu, J.; Xu, Y.; Song, Y.; Liu, Y. Reversible Formation of g-C3N4 3D Hydrogels through Ionic Liquid Activation: Gelation Behavior and Room-Temperature Gas-Sensing Properties. *Adv. Funct. Mater.* **2017,** *27* (22), 1700653.
(291) Huang, X.; Wang, J.; Li, T.; Wang, J.; Xu, M.; Yu, W.; El Abed, A.; Zhang, X. Review on optofluidic microreactors for artificial photosynthesis. *Beilstein journal of nanotechnology* **2018,** *9*, 30.
(292) Han, P.; Martens, W.; Waclawik, E. R.; Sarina, S.; Zhu, H. Metal Nanoparticle Photocatalysts: Synthesis, Characterization, and Application. *Part. Part. Syst. Charact.* **2018,** *35* (6), 1700489.
(293) Liu, G.; Du, K.; Xu, J.; Chen, G.; Gu, M.; Yang, C.; Wang, K.; Jakobsen, H. Plasmon-dominated photoelectrodes for solar water splitting. *J. Mater. Chem. A* **2017,** *5* (9), 4233.
(294) Eftekhari, A.; Babu, V. J.; Ramakrishna, S. Photoelectrode nanomaterials for photoelectrochemical water splitting. *Int. J. Hydrogen Energy* **2017,** *42* (16), 11078.
(295) Lan, Z. A.; Fang, Y.; Zhang, Y.; Wang, X. Photocatalytic Oxygen Evolution from Functional Triazine-Based Polymers with Tunable Band Structures. *Angew. Chem.* **2018,** *130* (2), 479.
(296) Yu, H.; Shi, R.; Zhao, Y.; Bian, T.; Zhao, Y.; Zhou, C.; Waterhouse, G. I.; Wu, L. Z.; Tung, C. H.; Zhang, T. Alkali-Assisted Synthesis of Nitrogen Deficient Graphitic Carbon Nitride with Tunable Band Structures for Efficient Visible-Light-Driven Hydrogen Evolution. *Adv. Mater.* **2017,** *29* (16), 1605148.
(297) Liu, C.; Huang, H.; Cui, W.; Dong, F.; Zhang, Y. Band structure engineering and efficient charge transport in oxygen substituted g-C3N4 for superior photocatalytic hydrogen evolution. *Appl. Catal. B* **2018,** *230*, 115.
(298) Gao, L.-F.; Wen, T.; Xu, J.-Y.; Zhai, X.-P.; Zhao, M.; Hu, G.-W.; Chen, P.; Wang, Q.; Zhang, H.-L. Iron-doped carbon nitride-type polymers as homogeneous organocatalysts for visible light-driven hydrogen evolution. *ACS Appl. Mater. Interfaces* **2015,** *8* (1), 617.
(299) Zhang, J.; Zhang, G.; Chen, X.; Lin, S.; Möhlmann, L.; Dołęga, G.; Lipner, G.; Antonietti, M.; Blechert, S.; Wang, X. Co-monomer control of carbon nitride semiconductors to optimize hydrogen evolution with visible light. *Angew. Chem. Int. Ed.* **2012,** *51* (13), 3183.
(300) Yang, P.; Wang, R.; Zhou, M.; Wang, X. Photochemical Construction of Carbonitride Superstructures for Red-Light Redox Catalysis. *Angew. Chem. Int. Ed.* **2018. DOI: https://doi.org/10.1002/anie.201804996/**.
(301) Wang, J.; Guan, Z.; Huang, J.; Li, Q.; Yang, J. Enhanced photocatalytic mechanism for the hybrid gC 3 N 4/MoS 2 nanocomposite. *J. Mater. Chem. A* **2014,** *2* (21), 7960.
(302) Hou, Y.; Wen, Z.; Cui, S.; Guo, X.; Chen, J. Constructing 2D Porous Graphitic C3N4 Nanosheets/Nitrogen-Doped Graphene/Layered MoS2 Ternary Nanojunction with Enhanced Photoelectrochemical Activity. *Adv. Mater.* **2013,** *25* (43), 6291.
(303) Peng, G.; Xing, L.; Barrio, J.; Volokh, M.; Shalom, M. A General Synthesis of Porous Carbon Nitride Films with Tunable Surface Area and Photophysical Properties. *Angew. Chem. Int. Ed.* **2018,** *57* (5), 1186.

(304) Safaei, J.; Rosli, N. N.; Mohamad Noh, M. F.; Mohamed, N. A.; Ibrahim, M. A.; Mat Teridi, M. A. Low temperature fabrication of transparent conductive electrode with high ultraviolet transmittance down to wavelength of 250 nm. *Phys. Status Solidi RRL* **2018. DOI: https://doi.org/10.1002/pssr.201800441.**